

**TRANSPORT AND ULTRAFAST TERAHERTZ  
EMISSION STUDIES ON EPITAXIAL  $\text{Co}_2\text{MnGe}$   
THIN FILMS**

**EKTA YADAV**



**DEPARTMENT OF PHYSICS  
INDIAN INSTITUTE OF TECHNOLOGY DELHI  
JULY 2025**



**TRANSPORT AND ULTRAFAST TERAHERTZ  
EMISSION STUDIES ON EPITAXIAL  $\text{Co}_2\text{MnGe}$   
THIN FILMS**

*by*

**EKTA YADAV**

Department of Physics

Submitted

in the fulfilment of the requirements of the degree of Doctor of Philosophy

to the



**INDIAN INSTITUTE OF TECHNOLOGY**

**DELHI**

**July 2025**

*Dedicated*  
*to*  
*My Parents and Teachers*

# CERTIFICATE

This is to certify that the thesis entitled, "**Transport and Ultrafast Terahertz Emission Studies on Epitaxial Co<sub>2</sub>MnGe Thin Films**", being submitted by **Ms. Ekta Yadav** to the Department of Physics, Indian Institute of Technology Delhi, New Delhi, for the award of degree of **Doctor of Philosophy** in Physics is a record of bonafide research work carried out by her under my supervision and guidance. She has fulfilled the requirements for submission of the thesis, which to the best of my knowledge has reached the requisite standard.

The results contained in the thesis have not been submitted in part or full to any other University or Institute for the award of any degree or diploma.

**Prof. Sunil Kumar**

Associate Professor

Department of Physics

Indian Institute of Technology Delhi

New Delhi 110016, India



# ACKNOWLEDGEMENT

---

I would like to express my sincere gratitude to my thesis advisor, Prof. Sunil Kumar, for providing me the opportunity to pursue my doctoral research under his guidance. I greatly appreciate his unwavering support and encouragement throughout my studies. His profound expertise, insightful guidance, and thoughtful approach have played a pivotal role in shaping my research skills and fostering a constructive academic mindset. His continuous inspiration has been invaluable to me during the course of my doctoral journey.

I want to thank the members of my SRC committee, Prof. Sujeet Chaudhary, Prof. B. K. Mani, and Prof. Satyananda Kar, for their fruitful discussions and constructive feedback regarding my research. I would also like to acknowledge the Department of Physics at IIT Delhi, for providing all the facilities for my research activities and travel grants. I acknowledge the Central Research Facility (CRF) and Nanoscale Research Facility (NRF) at IIT Delhi, and their scientific staff members, for their assistance with various measurements related to my research. I gratefully acknowledge the University Grant Commission (UGC), Government of India, for providing me a research fellowship.

My sincere thanks to all my labmates Dr. Madhu Bochalya, Dr. Arvind Singh, Dr. Anand Nivedan, Dr. Sandeep Kumar, Dr. Monu Kinha, Neetesh Dhakar, Satakshi Gupta, Shiwangi Puri, Prateek Nautiyal, Sandeep Jakhar, and Akanksha Yadav for their help and support during my PhD days. Thanks to my friends Nishu, Sumreti, Sushmita, Shruti, Nikita, Deepesh, Richa, Chinmay, Manoj, Arun, Arpita, Pankaj, Indraneel, Vireshwar, and Nakul, who made my stay on campus a pleasant and memorable experience. I am also grateful to my childhood friends Priyanka, Arshma, Farheen, and Rahul for their unconditional support and for the great times we shared.

I want to express my sincere respect and heartfelt gratitude to my family, whose support and understanding have always given me strength to move forward. I thank my brother Vikas for his love and support. I am filled with boundless love and appreciation

for my father and mother, whose unwavering support and sacrifices have been the foundation of my journey.

Above all, I bow my head in gratitude to God, whose blessings, guidance, and grace have given me strength, perseverance, and faith throughout this journey.

Lastly, I would like to express my sincere thanks to everyone who supported me directly or indirectly during my PhD journey.

**Ekta Yadav**

# Abstract

---

Ferromagnetic Co-based full Heusler alloys, known for high Curie temperature, tunable electronic properties, and half-metallicity. Recently, some of these Co-based ferromagnetic full Heusler alloys have been predicted to host Weyl fermions in their electronic band structure. The Berry curvature associated with this non-trivial topological state exhibit exotic phenomena, such as the anomalous Hall effect (AHE), Fermi arc surface states, and other Berry curvature-induced effects. Among these compounds,  $\text{Co}_2\text{MnGe}$ , which crystallizes in the  $Fm\bar{3}m$  space group, exhibits half-metallicity, high spin polarization and Weyl nodes in its band structure. These properties make  $\text{Co}_2\text{MnGe}$  an excellent candidate not only for spin-based electronic devices but also for THz emitter. Optimizing its growth conditions and investigating its transport and ultrafast optical response under various external conditions are the primary focus of this thesis.

The research work in the thesis is categorized into two sections, focusing on the structural, magnetotransport, and ultrafast Terahertz (THz) emission studies of  $\text{Co}_2\text{MnGe}$ . The first section focuses on the growth and fundamental characterization of  $\text{Co}_2\text{MnGe}$  thin films, which were deposited on MgO substrates using the pulsed laser deposition (PLD) technique. Structural characterization reveals that the films are epitaxial with B2 ordering, while transport measurements confirm their metallic behaviour. Further investigation using AHE measurements indicates that intrinsic mechanisms dominate over extrinsic contributions, implying a nonzero Berry curvature, which is a characteristic signature of Weyl semimetals. These findings suggest that  $\text{Co}_2\text{MnGe}$  is a candidate Weyl material, motivating further exploration of its topological and spintronic properties.

The second section focuses on ultrafast processes in  $\text{Co}_2\text{MnGe}$  thin films using THz time-domain spectroscopy, a powerful technique for probing topological states and spintronic phenomena. This section is driven by three key objectives. First, it is demonstrated that  $\text{Co}_2\text{MnGe}$  thin films emit THz radiation upon femtosecond laser excitation, with emission efficiency and topological properties being highly sensitive to growth temperature. To further understand the behaviour of the films, temperature,

wavelength, and polarization-dependent THz emission measurements were performed on an optimized film, providing insights into the influence of external parameters. Finally, we analysed the ultrafast photocurrent response in the surface as well as the bulk of  $\text{Co}_2\text{MnGe}$  thin film and investigated underlying linear and nonlinear mechanisms governing these responses at both room and low temperature (15K). The overall study presented in this thesis advances our fundamental understanding of  $\text{Co}_2\text{MnGe}$  ferromagnetic full Heusler alloy through rigorous analysis of its growth, structural characterization, and by investigating their topological characteristics via magnetotransport and ultrafast THz emission studies.

# सार

फेरोमैग्नेटिक Co-आधारित पूर्ण हेस्लर मिश्रधातु, उच्च क्यूरी तापमान, ट्यूनेबल इलेक्ट्रॉनिक गुणों और अर्ध-धात्विकता के लिए जाने जाते हैं। हाल ही में, इनमें से कुछ Co-आधारित फेरोमैग्नेटिक पूर्ण हेस्लर मिश्रधातुओं में उनके इलेक्ट्रॉनिक बैंड संरचना में वेइल फर्मियन की मेजबानी करने की भविष्यवाणी की गई है। इस गैर-तुच्छ टोपोलॉजिकल स्थिति से जुड़ी बेरी वक्रता असामान्य घटनाओं को प्रदर्शित करती है, जैसे कि असामान्य हॉल प्रभाव (AHE), फर्मी आर्क सतह की स्थिति और अन्य बेरी वक्रता-प्रेरित प्रभाव। इन यौगिकों में,  $\text{Co}_2\text{MnGe}$ , जो  $Fm\bar{3}m$  स्पेस ग्रुप में क्रिस्टलीकृत होता है, अपनी बैंड संरचना में अर्ध-धात्विकता, उच्च स्पिन ध्रुवीकरण और वेइल नोड्स प्रदर्शित करता है। ये गुण  $\text{Co}_2\text{MnGe}$  को न केवल स्पिन-आधारित इलेक्ट्रॉनिक उपकरणों के लिए बल्कि THz एमिटर के लिए भी एक उत्कृष्ट उम्मीदवार बनाते हैं। इसकी वृद्धि की स्थितियों को अनुकूलित करना और विभिन्न बाहरी स्थितियों के तहत इसके परिवहन और अल्ट्राफास्ट ऑप्टिकल प्रतिक्रिया की जांच करना इस थीसिस का प्राथमिक फोकस है।

थीसिस में शोध कार्य को दो खंडों में वर्गीकृत किया गया है, जो  $\text{Co}_2\text{MnGe}$  के संरचनात्मक, मैग्नेटोट्रांसपोर्ट और अल्ट्राफास्ट टेराहर्ट्ज़ (THz) उत्सर्जन अध्ययनों पर केंद्रित है। पहला खंड  $\text{Co}_2\text{MnGe}$  पतली फिल्मों की वृद्धि और मौलिक लक्षण वर्णन पर केंद्रित है, जिन्हें पल्स लेजर डिपोजिशन (PLD) तकनीक का उपयोग करके MgO सबस्ट्रेट पर जमा किया गया था। संरचनात्मक लक्षण वर्णन से पता चलता है कि फिल्में B2 ऑर्डरिंग के साथ एपिटैक्सियल हैं, जबकि परिवहन माप उनके धात्विक व्यवहार की पुष्टि करते हैं। AHE माप का उपयोग करके आगे की जांच से संकेत मिलता है कि आंतरिक तंत्र बाहरी योगदानों पर हावी है, जिसका अर्थ है एक गैर-शून्य बेरी वक्रता, जो वेइल सेमीमेटल्स का एक विशिष्ट हस्ताक्षर है। ये निष्कर्ष बताते हैं कि  $\text{Co}_2\text{MnGe}$  एक संभावित वेइल सामग्री है, जो इसके टोपोलॉजिकल और स्पिनट्रॉनिक गुणों की आगे की खोज को प्रेरित करती है।

दूसरा खंड THz टाइम-डोमेन स्पेक्ट्रोस्कोपी का उपयोग करके  $\text{Co}_2\text{MnGe}$  पतली फिल्मों में अल्ट्राफास्ट प्रक्रियाओं पर केंद्रित है, जो टोपोलॉजिकल अवस्थाओं और स्पिनट्रॉनिक घटनाओं की जांच के लिए एक शक्तिशाली तकनीक है। यह खंड तीन प्रमुख उद्देश्यों से प्रेरित है। सबसे

पहले, यह प्रदर्शित किया गया है कि  $\text{Co}_2\text{MnGe}$  पतली फिल्मों में फेमटोसेकंड लेजर उत्तेजना पर THz विकिरण उत्सर्जित करती हैं, जिसमें उत्सर्जन दक्षता और टोपोलॉजिकल गुण वृद्धि तापमान के प्रति अत्यधिक संवेदनशील होते हैं। फिल्मों के व्यवहार को और अधिक समझने के लिए, तापमान, तरंग दैर्घ्य और ध्रुवीकरण पर निर्भर THz उत्सर्जन माप एक अनुकूलित फिल्म पर किए गए, जो बाहरी मापदंडों के प्रभाव के बारे में जानकारी प्रदान करते हैं। अंत में, हमने सतह के साथ-साथ  $\text{Co}_2\text{MnGe}$  पतली फिल्म के थोक में अल्ट्राफास्ट फोटोकॉरंट प्रतिक्रिया का विश्लेषण किया और कमरे और कम तापमान (15K) दोनों पर इन प्रतिक्रियाओं को नियंत्रित करने वाले अंतर्निहित रैखिक और गैर-रैखिक तंत्रों की जांच की। इस थीसिस में प्रस्तुत समग्र अध्ययन  $\text{Co}_2\text{MnGe}$  फेरोमैग्नेटिक पूर्ण ह्यूस्लर मिश्रधातु के विकास, संरचनात्मक लक्षण वर्णन के कठोर विश्लेषण, तथा मैग्नेटोट्रांसपोर्ट और अल्ट्राफास्ट THz उत्सर्जन अध्ययनों के माध्यम से उनकी टोपोलॉजिकल विशेषताओं की जांच के माध्यम से हमारी मौलिक समझ को आगे बढ़ाता है।

# Table of contents

---

<i>Certificate</i>	i
<i>Acknowledgement</i>	iii
<i>Abstract (English)</i>	v
<i>Abstract (Hindi)</i>	vii
<i>Table of Contents</i>	ix
<i>List of Figures</i>	xiii
<i>List of Tables</i>	xxv
<i>List of abbreviations</i>	xxvii
<b>Chapter 1: Introduction</b>	<b>1</b>
1.1 Heusler compounds and spintronics	1
1.1.1 Classification of Heusler compounds	2
1.1.2 Full Heusler alloys and their properties	3
1.1.2.1 Crystal structure of full Heusler alloys	3
1.1.2.2 Origin of Half metallicity in full Heusler alloys	5
1.1.2.3 Magnetic moment of full Heusler alloys	8
1.1.2.4 Emergence of magnetic Weyl fermions in Co-based full Heusler alloys	9
1.2 Berry curvature induced properties of magnetic Weyl semimetal	10
1.3 Few potential applications of Co-based full Heusler alloys	15
1.4 Ultrafast THz emission from femtosecond laser excited ferromagnetic Heusler alloy thin films and their heterostructures	16

1.5	Choice of Co <sub>2</sub> MnGe Heusler compound	20
1.6	Motivation and objectives of the thesis	22
1.7	Organization of the thesis	24
	Bibliography	27
	<b>Chapter 2: Materials and Methods</b>	<b>35</b>
2.1	Thin film preparation and characterization	35
2.1.1	Pulsed laser deposition technique	35
2.1.2	Physical properties of thin films	37
2.1.2.1	Crystalline purity using X-ray diffraction	37
2.1.2.2	Thickness measurement using X-ray reflectivity	39
2.1.2.3	Surface morphology by atomic force microscopy	41
2.1.3	Electrical and magnetotransport measurements	43
2.1.3.1	Electric resistivity measurements	44
2.1.3.2	Anomalous Hall effect measurements	44
2.2	Ultrafast time-domain measurements at room temperature	45
2.2.1	Femtosecond laser source and pulse characteristics	46
2.2.1.1	Intensity autocorrelation	49
2.2.1.2	Interferometric Autocorrelation	51
2.2.1.3	Determination of Pulse Parameters	52
2.2.2	Overview of the THz time-domain spectroscopy	53
2.2.2.1	THz radiation and pulse generation	53
2.2.2.2	THz experimental setup	56
2.2.2.3	THz pulse detection by electro-optic sampling	57
2.2.3	Low temperature dependent THz time-domain experiments	59
	Bibliography	63

<b>Chapter 3: Anomalous Hall effect in pulsed laser deposited epitaxial Co<sub>2</sub>MnGe full Heusler alloy thin films.</b>	<b>65</b>
3.1 Thin film preparation and optimization	66
3.2 Film characterizations	67
3.2.1 Structural characterization by XRD and Phi scan measurement	67
3.2.2 Film thickness and surface morphology	69
3.3 Ferromagnetic behaviour of thin film	70
3.4 Temperature dependent electrical and magnetotransport measurements for Co <sub>2</sub> MnGe thin film	71
3.4.1 Longitudinal resistivity and magnetoresistance	71
3.4.2 Anomalous Hall effect in epitaxial Co <sub>2</sub> MnGe thin films	73
3.5 Summary	78
Bibliography	79
<b>Chapter 4: THz probing of non-trivial topological states in Co<sub>2</sub>MnGe Heusler alloy thin films</b>	<b>83</b>
4.1 Sample preparation and optimization	84
4.2 Effect of substrate temperature on the crystallinity and MH behaviour of Co <sub>2</sub> MnGe thin films	85
4.3 Substrate temperature dependent THz pulse emission	86
4.4 THz pulse emission under external magnetic field	88
4.5 Excitation pump helicity dependent photocurrent as the source for THz radiation	91
4.6 Summary	94
Bibliography	95
<b>Chapter 5: Excitation photon energy and sample temperature dependence of THz emission from Co<sub>2</sub>MnGe epitaxial thin film</b>	<b>99</b>

5.1	Fluence dependent THz generation from thin film	101
5.2	Magnetic field free THz emission	102
5.3	Temperature dependent THz emission	103
5.4	Wavelength dependent THz pulse emission	105
5.5	Polarization dependent THz emission	108
5.6	Comparison of THz emission between Co <sub>2</sub> MnGe and Fe/Pt thin film	109
5.7	Summary	111
	Bibliography	113
	<b>Chapter 6: Tunable helicity dependent photocurrent response in topological Co<sub>2</sub>MnGe epitaxial thin film</b>	<b>117</b>
6.1	Film characterization	118
6.2	Helicity-dependent THz pulse emission	119
6.3	Ultrafast photocurrent response as a function of pump fluence	123
6.4	Separating fluence-dependent surface and bulk contributions to THz emission at 300 K and 15 K	130
6.5	Summary	131
	Bibliography	133
	<b>Chapter 7: Conclusions and Future Scope</b>	<b>135</b>
7.1	Conclusion	135
7.2	Future scope of the thesis work	136
	<b>List of Publications</b>	<b>139</b>
	<b>Author's Biodata</b>	<b>141</b>

# LIST OF FIGURES

---

Figure No.	Figure Caption	Page No.
1.1	An illustrative representation of the electronic density of states (DOS) for different materials. (a) metal, (b) half-metal, (c) semiconductor, (d) topological insulator, and (e) Weyl semimetal.	2
1.2	Schematic representation of different types of Heusler compound structures (a) half-Heusler (structure C1 <sub>b</sub> ), (b) full-Heusler (structure L2 <sub>1</sub> ), (c) inverse-Heusler, and (d) quaternary Heusler compounds.	3
1.3	Schematic representation of the X <sub>2</sub> YZ full Heusler alloy unit cell, where different coloured circles indicate the position of atoms, namely the X (red), Y (blue), and Z (green). The structural configurations are classified as (a) L2 <sub>1</sub> (perfectly ordered), (b) B2 (partially ordered), and (c) A2 (completely disordered). The unit cell adopts a face-centred cubic (fcc) lattice with four atomic positions: X at (0,0,0) and (1/2, 1/2, 1/2), Y at (1/4, 1/4, 1/4), and Z at (3/4, 3/4, 3/4), as defined by Wyckoff coordinates.	4
1.4	Total density of states (DOS) for Co <sub>2</sub> MnZ Heusler compounds, where Z represents Al, Si, Ge, and Sn. (Adapted from ref.[22])	6
1.5	Possible hybridizations of spin-down orbitals located at different lattice sites in the Co <sub>2</sub> MnGe compound. The analysis of the full Heusler alloy properties is carried out by first examining the hybridization between the two distinct Co atoms (a), followed by the hybridization involving the Mn atom (b). Figure redrawn from ref.[22].	7

<b>1.6</b>	Total magnetic moment of full Heusler alloys calculated based on the total number of valence electrons, following the Slater-Pauling rule (Adapted from ref. [22]).	9
<b>1.7</b>	Different topological phases observed in Heusler compounds. (a) A half-Heusler compound demonstrating a topological insulating state and (b) full-Heusler compound exhibiting a Weyl semimetal phase. Adapted from [2, 29].	10
<b>1.8</b>	Schematic representation of the nontrivial band structure and Berry curvature in a topological semimetal. (a) Energy dispersion near the Weyl points, where the conduction and valence bands touch, forming a linear crossing in momentum space. (b) Vector field illustration of Berry curvature in k-space, where the red and blue nodes represent Weyl points with opposite chirality, acting respectively as sources and sinks of Berry curvature. Adapted from ref. [43].	11
<b>1.9</b>	Schematic of spin-orbit interaction: In the reference frame of a moving electron (depicted as a green sphere with velocity $V_e$ ), the electric field $E$ generated by the nucleus (represented by a red sphere) is perceived as an effective magnetic field ( $B_{\text{eff}}$ ). The coupling between the electron's intrinsic spin and this effective magnetic field constitutes the spin-orbit interaction phenomenon. Figure redrawn from ref. [44].	12
<b>1.10</b>	Intrinsic and extrinsic mechanisms contributing to the AHE. Schematic representation of (a) intrinsic deflection driven by Berry curvature. (b) skew scattering is caused by asymmetrical impurity scattering, and (c) the side jump mechanism results from spin-orbit interaction near magnetic impurity potentials.	14
<b>1.11</b>	Ultrafast magnetization dynamics and THz emission from a ferromagnetic thin film. (a) Rapid demagnetization in ferromagnetic Ni triggered by a 60 fs laser pulse, illustrating a sharp decrease in	17

remanence followed by partial recovery over time. (b) THz radiation generated due to the ultrafast demagnetization process in a thick Ni single layer, where curve (1) shows the measured THz signal and curve (2) represents the corresponding simulated magnetization response. Adapted from Ref. [85, 92].

<b>1.12</b>	THz emission from Co <sub>2</sub> FeAl (CFA)/Pt bilayer structures. The green vertical line marks the THz emission amplitude for reference Pt (3)/Fe (2)/MgO spintronic emitter, used as a benchmark. This comparison highlights the efficiency of the CFA/Pt heterostructures for ultrafast spintronic THz emission.	19
<b>1.13</b>	Helicity-dependent THz emission from an 80 nm Co <sub>2</sub> MnGa thin film. The THz field amplitude is plotted as a function of the quarter-wave plate (QWP) angle $\alpha$ . The variation of the THz amplitude follows a sinusoidal dependence on the QWP angle. Adapted from Ref. [84].	20
<b>1.14</b>	Density of states (DOS) for Co <sub>2</sub> MnGe in two different structural phases. (a) Represents the DOS in the B2 phase, where half-metallicity is observed, while (b) corresponds to the A2 phase, which lacks half-metallic behaviour. The contributions from Co, Mn, and Ge atoms to the electronic states are indicated in the legend. (Adapted from ref. [119]).	21
<b>2.1</b>	(a) Pictorial representation of the PLD system used in this study. (b) The schematic diagram for the PLD process and vacuum chamber.	36
<b>2.2</b>	(a) Real time image of plume taken during pre-deposition. (b) Image of the composite Co <sub>2</sub> MnGe target used to produce thin films in the thesis.	37
<b>2.3</b>	Schematic showing diffraction of X-rays from the atomic planes of a typical crystalline material. d shows the lattice spacing distance.	38
<b>2.4</b>	(a) Schematic diagram of an X-ray diffractometer, showing the X-ray	39

	source, detector, and the paths of both incident and diffracted X-rays.	
	(b) Setup of the X-ray diffractometer for grazing incidence X-ray diffraction (GI-XRD), where the incident angle remains constant while the detector rotates to meet Bragg's condition.	
<b>2.5</b>	Different angles, indices, and wave vector directions of refraction, when X-rays are incident from the air on the surface of a sample with a refractive index less than 1.	40
<b>2.6</b>	A representative XRR spectra of Co <sub>2</sub> MnGe (30nm) thin film grown on Si (100) substrate showing the different information that can be obtained from XRR spectra.	41
<b>2.7</b>	Schematic diagram of the atomic force microscopy measurement setup.	43
<b>2.8</b>	Schematic diagram of the four-probe linear resistivity measurement configuration used for highly conducting thin film samples.	44
<b>2.9</b>	Schematic of the four-point probe Hall effect measurement geometry.	45
<b>2.10</b>	Schematic representation of the high-intensity femtosecond pulse generation mechanism using a Ti:Sapphire-based regenerative amplifier laser system (Astrella, available in our laboratory). The diagram illustrates the fundamental working principle of chirped pulse amplification. Initially, weak femtosecond pulses from the seed laser are temporally stretched in the stretcher. The first Pockels cell (PC1) selectively isolates a single pulse for subsequent amplification. Following amplification, the second Pockels cell (PC2) guides the amplified pulse to the compressor, where it is recompressed to yield high-intensity, transform-limited femtosecond pulses at the output.	48
<b>2.11</b>	(a) Schematic diagram of the intensity autocorrelation setup utilizing second-harmonic generation (SHG). The experimental arrangement employs a beam splitter, retroreflector, and mirrors to direct the laser	50

pulses in a non-collinear geometry toward the BBO (beta-barium borate) crystal. (c) A comparative illustration demonstrating the relationship between the Gaussian-shaped pulse and its corresponding autocorrelation trace. (b) Measured second-harmonic intensity autocorrelation signal of the amplifier output obtained in our laboratory. The recorded signal displays a full width at half maximum (FWHM) of approximately 128 fs, which corresponds to an estimated pulse duration of ~90 fs after applying the appropriate deconvolution factor.

- 2.12** (a) Schematic representation of the PSAC technique utilizing a Michelson interferometer configuration. (b) The output signal from the autocorrelator (Avesta, model: AA-10DD-12PS), indicating a measured FWHM pulse duration of approximately 38 fs. 51
- 2.13** Schematic illustration of the electromagnetic spectrum, encompassing various regions from radio waves to X-rays. The terahertz (THz) range is highlighted to emphasize the transition between electronics and photonics. Essential parameters, including frequency, wavelength, wavenumber, period, photon energy, and the corresponding temperature values, are presented for reference. Recreated from reference [14]. 54
- 2.14** Illustration of the diverse applications of THz radiation across multiple fields. The top-left schematic depicts the use of THz spectroscopy in security and non-destructive testing, while the remaining schematics highlight applications in areas such as medical diagnostics, THz spectroscopy in fundamental scientific research, and wireless communication networks. Adapted from references [17, 19, 20]. 55
- 2.15** Schematic illustration of various mechanisms responsible for THz pulse emission from single layer of Heusler alloys upon fs optical excitation. The diagram shows four principal mechanisms: (a) 56

ultrafast demagnetization (b) photon drag effect (c) optical rectification, and (d) circular photogalvanic effect. Adapted from references [25-27].

<b>2.16</b>	Schematic of the time-domain THz emission spectroscopy setup in reflection geometry. The system consists of a Ti:Sapphire based regenerative amplifier, a beam splitter to divide the laser output, a motorized stage for delaying the probe pulse, and off-axis parabolic mirrors to collect the emitted THz pulses. The detection utilizes an electro-optic sampling technique incorporating a quarter-wave plate, a Wollaston prism, and a balanced photodiode.	57
<b>2.17</b>	Experimental setup for THz waveform detection using the electro-optic sampling method. A (110)-oriented ZnTe crystal is used for detection. The probe polarizations, both in the absence and presence of the THz field, are illustrated before and after the QWP and WP. The QWP and WP are employed to separate the orthogonally polarized components of the probe pulse's electric fields. The intensity difference between ( $\Delta I$ ) between PD1 and PD2 is measured using a lock-in amplifier.	58
<b>2.18</b>	Schematic diagram of the time-domain THz emission measurement setup, which is integrated with a low-temperature optical cryostat to enable temperature-dependent studies of the sample, ranging from room temperature to 5 Kelvin (K).	60
<b>2.19</b>	Schematic diagram of the optical cryostat system: Janis SHI-4-2-XG closed-cycle helium cryostat. (a) Cryostat vibration isolation design featuring rubber bellows and silicone gel pads, (b) Lake Shore 335 temperature controller, (c) Turbomolecular pump, and (d) Cryostat chamber with optical and THz windows.	61
<b>2.20</b>	Co <sub>2</sub> MnGe thin film sample mounted on a gold-plated copper holder.	62
<b>3.1</b>	Crystal structure of PLD grown (100)-oriented epitaxial Co <sub>2</sub> MnGe	69

- thin film on MgO (100) substrate. (a) Out-of-plane XRD patterns, (b)  $\phi$ -scan data (along (220) planes) of the Co<sub>2</sub>MnGe film (top) and the MgO substrate (bottom). (c) Schematic diagram of the B2-ordered crystal structure (left) and top view of Co<sub>2</sub>MnGe unit cell over MgO lattice (right).
- 3.2** (a) X-ray reflectivity data (blue circles) and fit (red line) for the 45 nm thick Co<sub>2</sub>MnGe film. The inset shows an AFM image of area ( $2 \times 2 \mu\text{m}^2$ ) with a scale bar of 400 nm. (b) Shows the mixed elemental mapping of Co, Mn, and Ge elements. 70
- 3.3** Magnetic properties of the epitaxial Co<sub>2</sub>MnGe film on MgO (100) substrate. (a) In-plane magnetic hysteresis (M-H) loops at various sample temperatures, plotted after subtracting the diamagnetic contribution of the MgO substrate. The inset shows the zoomed-in view of M-H loop for determining coercivity variation with temperature. (b) Variation of coercivity with the sample temperature. 71
- 3.4** Electrical properties of the epitaxial Co<sub>2</sub>MnGe film on MgO (100) substrate. (a) Temperature-dependent (2-300 K) longitudinal resistivity and model fitting of the data in two distinct regions: low-T region of 2-80 K (region-I) and high-T region of 80-300 K (region-II). (b) Magnetoresistance data taken at different temperatures by varying the magnetic field perpendicular to the applied current. Inset: Schematic of the sample and measurement geometry. 72
- 3.5** AHE in epitaxial Co<sub>2</sub>MnGe thin film. (a) Magnetic field-dependent Hall resistivity,  $\rho_{xy}$  at various sample temperatures as indicated. The solid line represents linear fit to the data at high field values and extrapolated to zero field to infer the anomalous Hall resistivity,  $\rho_{xy}^{\text{AHE}}$ . Inset: experimental geometry adopted for these measurements. (b) Ordinary Hall coefficient as a function of the sample temperature. Inset: calculated values of the carrier concentration. (c) Anomalous 75

- Hall resistivity,  $\rho_{xy}^{\text{AHE}}$  as a function of temperature. (d) Field-dependent Hall conductivity at various temperatures. Inset represents the linear fitting of the data at high fields and extrapolated to zero field to infer the anomalous Hall conductivity,  $\sigma_{xy}^{\text{AHE}}$ .
- 3.6** (a) Variation of  $\rho_{xy}^{\text{AHE}}$  with  $\rho_{xx}^2$ , and (b) variation of  $\sigma_{xy}^{\text{AHE}}$  with  $\sigma_{xx}^2$ . 77  
Solid lines are fits obtained using the TYJ model as discussed in the text. (c) Temperature-dependence of  $\sigma_{xy}^{\text{AHE}}$ . (d) Temperature-dependence of scaling coefficient,  $S_H$ .
- 4.1** Crystalline and magnetic properties of CMG-500, CMG-600, and 85  
CMG-650 thin films. (a) X-ray diffraction patterns of the different CMG thin films and the MgO substrate. Vertical dashed lines mark the positions of the highly oriented (200) and (400) crystal planes in the CMG lattice. (b) AFM topography images of the CMG films in an area of  $5 \times 5 \mu\text{m}^2$ . (c) Magnetic hysteresis loops of CMG thin films obtained for in-plane external magnetic field. (d) Saturation magnetization,  $M_S$  and coercive field,  $H_C$  as a function of substrate temperature.
- 4.2** THz emission spectroscopy measurements on CMG-500, CMG-600, 87  
and CMG-650 thin films. (a) Schematic diagram of the experimental setup depicting the optical fs laser pulse excitation, THz emission and pick-off in the reflection geometry. Proper rotation ( $\alpha$ ) of the quarter wave plate QWP ensures different polarization states of the excitation light. (b) Typical time-domain THz signals and (c) the Fourier spectra spanning over above 3 THz bandwidth obtained with the linearly polarized ( $\alpha = 0^\circ$ ) excitation pulses.
- 4.3** Magnetic field direction dependent THz emission from CMG-500, 89  
CMG-600 and CMG-650 thin films. (a) THz signals emitted by the photoexcited films under oppositely directed external static magnetic field  $\pm B = \pm 2000$  Oe. (b) Spin-dependent (odd) and spin-independent

- (even) components of the THz signals. See text for more details.
- 4.4** Excitation pulse helicity dependent THz emission from CMG-500, CMG-600, and CMG-650 films. The experimentally recorded THz waveforms obtained under excitation by LP ( $\alpha = 0^\circ$ ), RCP ( $\alpha = 45^\circ$ ) and LCP ( $\alpha = -45^\circ$ ) light. The data for different samples are shifted to the horizontal axis for better clarity. The THz peak-to-peak amplitude indicated by  $E_{PP}$ . 91
- 4.5** (a) Variation of the magnitude of the peak-to-peak amplitude of the THz signals ( $E_{PP}$ ) as indicated in **Fig. 4.4** with respect to the substrate temperature  $T_s$ . (b) Variation of the CPGE induced contribution to the THz emission with respect to the substrate temperature  $T_s$ . 92
- 5.1** Pump fluence dependence of the THz emission from  $\text{Co}_2\text{MnGe}$  thin film under the linear polarized laser excitation and an applied magnetic field ( $+B = 2000$  Oe). (a) Time-domain THz waveforms measured at different pump fluences ranging from  $80 \mu\text{J}/\text{cm}^2$  to  $810 \mu\text{J}/\text{cm}^2$ . Inset shows the fast Fourier transform (FFT) spectra of the directly measured THz waveforms. (b) The extracted THz peak-to-peak amplitude ( $E_{PP}$ ) as a function of pump fluence. The solid black line represents a linear fit to the data, indicating overall linear dependence. 102
- 5.2** THz emission from  $\text{Co}_2\text{MnGe}$  thin film under different magnetic field conditions. (a) Time domain THz signal measured with and without an applied magnetic field ( $+B = 2000$  Oe) and  $0B$ ). (b) Peak to peak THz field amplitude ( $E_{PP}$ ) as a function of pump fluence for both field conditions. 103
- 5.3** Temperature-dependent THz emission from  $\text{Co}_2\text{MnGe}$  thin film. (a) Time domain THz signals measured at different sample temperatures ( $18$  K,  $60$  K,  $110$  K, and  $300$  K), under a pump fluence of  $800 \mu\text{J}/\text{cm}^2$ . 104

- These temporal signals are horizontally shifted for clarity. (b) Peak to peak THz amplitude ( $E_{PP}$ ) as a function of temperature.
- 5.4** Transport and magnetic measurements on  $\text{Co}_2\text{MnGe}$  thin film. (a) 105  
 Temperature-dependent longitudinal resistivity indicating the metallic nature of the film. (b) Magnetic hysteresis loops measured at different film temperatures. (c) Comparison of normalized THz peak amplitude (black circles) and extracted normalized magnetization (red solid line) from (b) as a function of temperature.
- 5.5** Wavelength-dependent THz emission from  $\text{Co}_2\text{MnGe}$  thin film. (a) 106  
 Time domain THz signals measured at different excitation wavelengths (640 nm, 800 nm, and 1200 nm), at a fixed pump fluence of  $80 \mu\text{J}/\text{cm}^2$ . (b) Peak-to-peak THz amplitude ( $E_{PP}$ ) as a function of excitation wavelength.
- 5.6** Wavelength-dependent THz emission and absorbance of  $\text{Co}_2\text{MnGe}$  107  
 thin film. The black curve with filled circles represents the extracted THz peak-to-peak electric field amplitude ( $E_{PP}$ ) measured at different excitation wavelengths (640 nm, 800 nm, and 1200 nm). The red curve shows the corresponding absorbance spectrum obtained from UV-Vis measurements.
- 5.7** THz emission from  $\text{Co}_2\text{MnGe}$  thin film under different linear pump 109  
 polarization angles. The THz peak amplitude is plotted as a function of the incident light polarization angle ( $\phi$ ). The blue curve represents the sinusoidal fitting, and the red dots are the experimental peak data. The arrows represent the polarization state of the incident light.
- 5.8** Comparison of THz emission from single-layer  $\text{Co}_2\text{MnGe}$  (30 nm) 111  
 and bilayer  $\text{Co}_2\text{MnGe}$  (30 nm)/Pt (3 nm) thin films. The insets show optical microscope images of the corresponding film surfaces.
- 6.1** Characterization of  $\text{Co}_2\text{MnGe}$  thin film. (a)  $\phi$ -scan of the film and the 119  
 MgO substrate along (220) planes. (b) AFM and (c) FESEM images

displaying the surface morphology of the film.

- 6.2** (a) Optical microscopy image of the sample with the schematic representation of the THz emission in the reflection geometry upon excitation of 800 nm ultrashort pump pulse with tunable pump polarization. (b) Transient THz waveforms with the inset showing the corresponding fast Fourier transform spectra from Co<sub>2</sub>MnGe thin film under the LP, RCP and LCP excitations, respectively. 120
- 6.3** Helicity-dependent THz emission observed in Co<sub>2</sub>MnGe thin film at 300K. (a) THz peak to peak amplitude,  $E_{PP}$  as a function of the QWP angle ( $\alpha$ ). Open circles and red lines represent the experimental data and fitting result by Eq. (6.1), respectively. (b)  $C \sin 2\alpha$  (red line),  $L_1 \sin 4\alpha$  (green line),  $L_2 \cos 4\alpha$  (blue line), and  $D$  (olive green line) as function of QWP angle ( $\alpha$ ) extracted using Eq. (6.1) from (a). For clarity, the large  $D$  component is scaled down to  $D/2$  in the plot. The inset shows the magnitudes of the  $C$ ,  $L_1$ ,  $L_2$  and  $D$  obtained by fitting of the experimental data. 122
- 6.4** (a-e) Peak-to-peak amplitude of emitted THz radiation as a function of QWP angles for different pump fluences at 300K. The open circle and solid lines represent experimental data and fitting data using Eq. (6.1), respectively. 123
- 6.5** (a)  $C \sin(2\alpha)$ , (b)  $L_1 \sin(4\alpha)$ , (c)  $L_2 \cos(4\alpha)$ , and (d) the polarization-independent offset  $D$ , all extracted by fitting the helicity-dependent THz emission data (Fig. 6.4) using Eq. (6.1), for different pump fluences at 300 K. 124
- 6.6** (a-c) Variation of the extracted fitting parameters  $C$ ,  $L_2$ , and  $D$  as a function of excitation pump fluence at 300 K. The parameters are extracted by fitting Eq. (6.1) with THz peak to peak experimental data at different pump fluences. The filled circle is the experimental data, and the solid lines represent the fitting. 125

- 6.7** Helicity-dependent THz emission observed in Co<sub>2</sub>MnGe thin film at 15 K for various excitation pump fluences. (a) The THz peak to peak amplitude,  $E_{PP}$  as a function of QWP angle ( $\alpha$ ). The red solid line is the best fit with Eq. (6.1). (b)  $C \sin 2\alpha$  (red line),  $L_1 \sin 4\alpha$  (green line),  $L_2 \cos 4\alpha$  (blue line), and  $D$  (olive green line) as function of QWP angle ( $\alpha$ ) extracted using Eq. (6.1) from (a). The large component  $D$  is scaled down to  $D/2$  for clarity in the plot. The inset shows the magnitude of the  $C$ ,  $L_1$ ,  $L_2$  and  $D$  obtained by fitting of the experimental data. 126
- 6.8** (a-e) Peak-to-peak amplitude of emitted THz radiation as a function of QWP angles for different pump fluences at 15 K. The filled circle and solid lines represent experimental data and fitting data using Eq. (6.1), respectively. 127
- 6.9** (a)  $C \sin(2\alpha)$ , (b)  $L_1 \sin(4\alpha)$ , (c)  $L_2 \cos(4\alpha)$ , and (d) the polarization-independent offset  $D$ , all extracted by fitting the helicity-dependent THz emission data (Fig. 6.8) using Eq. (6.1), for different pump fluences at 15 K. 128
- 6.10** (a-c) Variation of the extracted fitting parameters  $C$ ,  $L_2$  and  $D$  as a function of excitation pump fluence at 15 K. The parameters are extracted by fitting Eq. (6.1) with THz peak to peak experimental data at different pump fluences. The filled circle is the experimental data, and the solid lines represent the fitting. 129
- 6.11** Extracted peak-to-peak THz amplitude,  $E_{PP}$  values for the parameters  $C$ ,  $L_2$ , and  $D/2$ , obtained from fitting, measured at 300 K and 15 K with a fixed pump fluence of  $810 \mu\text{J}/\text{cm}^2$ . The red pattern bars correspond to 300 K, and the blue pattern bars represent 15 K. 130
- 6.12** Extracted peak to peak values of the THz amplitude contributed by both the bulk and surface states as a function of the pump fluence at 300 K and 15 K temperatures. The solid black and red lines 131

correspond to the fitting curves, while the filled and open circles denote the experimental data.

# LIST OF TABLES

---

<b>Table No.</b>	<b>Table Caption</b>	<b>Page No.</b>
4.1	Calculated spin-ordering dependent (odd) and spin-ordering independent (even) contributions to the THz emission from CMG films. The percentage contributions, i.e., $E_{\text{odd}}^{\text{THz}} / (E_{\text{even}}^{\text{THz}} + E_{\text{odd}}^{\text{THz}})$ and $E_{\text{even}}^{\text{THz}} / (E_{\text{even}}^{\text{THz}} + E_{\text{odd}}^{\text{THz}})$ , are provided inside the brackets.	90



# LIST OF ABBREVIATION

---

<b>ARPES</b>	Angle-resolved photoemission spectroscopy
<b>BS</b>	Beam splitter
<b>CPGE</b>	Circular photogalvanic effect
<b>EOS</b>	Electrooptic sampling
<b>WP</b>	Wollaston prism
<b>FFT</b>	Fast Fourier transform
<b>FWHM</b>	Full width at half maximum
<b>HHG</b>	High harmonic generation
<b>DOS</b>	Density of states
<b>IAC</b>	Intensity autocorrelation
<b>LCP</b>	Left circular polarization
<b>RCP</b>	Right circular polarization
<b>LP</b>	Linear polarization
<b>LPGE</b>	Linear photogalvanic effect
<b>MR</b>	Magnetoresistance
<b>OPA</b>	Optical parametric amplification
<b>OR</b>	Optical rectification
<b>HWP</b>	Half wave plate
<b>QWP</b>	Quarter wave plate
<b>PC</b>	Pockels cell
<b>PCS</b>	Photoconductive sampling
<b>SHG</b>	Second-harmonic generation
<b>PM</b>	Parabolic mirror
<b>PPMS</b>	Physical property measurement system
<b>MPMS</b>	Magnetic property measurement system
<b>PSAC</b>	Phase-sensitive autocorrelation

<b>AFM</b>	Atomic force microscopy
<b>XRR</b>	X-ray reflection
<b>THz</b>	Terahertz
<b>PLD</b>	Pulsed laser deposition
<b>ISHE</b>	Inverse spin Hall effect
<b>OHE</b>	Ordinary Hall effect
<b>SOC</b>	Spin orbit coupling
<b>AHE</b>	Anomalous Hall effect
<b>XRD</b>	X-ray diffraction
<b>WSM</b>	Weyl semimetal
<b>STE</b>	Spintronic THz emitter