



An-Najah National University
Faculty of Graduate Studies

**EXPLORING THE STRUCTURAL, MECHANICAL,
ELECTRONIC, MAGNETIC, OPTICAL AND
THERMOELECTRIC PROPERTIES OF
QUATERNARY HEUSLER ALLOYS
FeMnScGa, FeMnScAl AND FeMnCrGe**

By
Hasan Ali Hasan Masri

Supervisor
Prof. Mohammed Abu-Jafar

**This Dissertation is submitted in Partial Fulfillment of the Requirements for the
Degree of Ph.D in Physics, Faculty of Graduate Studies, An-Najah National
University, Nablus, Palestine.**

2025

EXPLORING THE STRUCTURAL, MECHANICAL, ELECTRONIC, MAGNETIC, OPTICAL AND THERMOELECTRIC PROPERTIES OF QUATERNARY HEUSLER ALLOYS FeMnScGa, FeMnScAl AND FeMnCrGe

**By
Hasan Ali Hasan Masri**

This Dissertation was Defended Successfully on 27/04/2025 and approved by

Prof. Mohammed Abu-Jafar

Supervisor

Prof. Mohammed Shabat

External Examiner

Prof. Mohammad Elsaid

Internal Examiner

Prof. Sami Jabr

Internal Examiner

Signature

Signature

Signature

Signature



An-Najah National University

Faculty of Graduate Studies

**EXPLORING THE STRUCTURAL, MECHANICAL,
ELECTRONIC, MAGNETIC, OPTICAL AND
THERMOELECTRIC PROPERTIES OF
QUATERNARY HEUSLER ALLOYS
FeMnScGa, FeMnScAl AND FeMnCrGe**

By

Hasan Ali Hasan Masri

Supervisor

Prof. Mohammed Abu-Jafar

In accordance with An-Najah National University Deans Council regulations for the award of Doctor of Philosophy, the following paper has been published after its extraction from the dissertation:

Masri, H. A., Abu-Jafar, M. S., Bouhemadou, A., & Baadji, N. (2025). Multifunctional Properties of FeMnScAl Quaternary Heusler Alloy: Insights into Spintronics, Photovoltaics, and Thermoelectric Applications. *The Journal of Physical Chemistry C*, 129(5), 2672-2690.

Masri, H. A., Abu-Jafar, M. S., Mohammad, N. F. A., & Essaoud, S. S. (2025). Extensive DFT study of FeMnCrGe quaternary Heusler alloy: structural, elastic, magnetic, optical and thermoelectric properties. *Optical and Quantum Electronics*, 57(2), 139.

Dedication

This work is dedicated to the cherished memory of my beloved parents, whose love, sacrifices, and unwavering support remain a constant source of inspiration. May this work be the image of that lasting impression they left on me and the principles they taught me with such enormous care. To my dear wife, whose love, patience, and encouragement proved my guiding light throughout. I am deeply grateful to my beloved sons and daughters, who infuse my life with joy, meaning, and inspiration. This work is a tribute to my parents' legacy, reflecting the dreams they held for me and inspiring my family to pursue their aspirations with unwavering determination and hope.

Acknowledgement

I wish to extend my sincere thanks to my supervisor, Prof. Dr. Mohammed Abu-Jafar, for his exceptional mentorship, enduring encouragement, and steadfast support. His expertise and insights have been crucial in determining both the direction and achievements of my research. I am also profoundly thankful to all the professors who contributed to my academic growth during my Ph.D. coursework. I am truly appreciative of all the professors whose knowledge and mentorship enhanced my academic experience throughout my Ph.D. research. I would especially like to thank Prof. Dr. Sami Al Jaber and Prof. Dr. Mohammad Elsaied for their unwavering support and lasting influence.

My appreciation goes to my son, Ali Masri, whose technical assistance has been instrumental in completing this work. I am profoundly thankful to my friends and colleagues for their cooperation, friendship, and insightful conversations that have profoundly impacted my academic journey. Your steadfast encouragement has made this experience both memorable and rewarding.

I am forever thankful to my family for their steadfast love, patience, and confidence in me. Their support has been the bedrock of my perseverance throughout this journey. I would like to express sincere thanks to all those people who contributed to this thesis in one way or another and contributed to the completion of this thesis. No matter how small each one's contribution may seem, it has been very important to me.

Declaration

I, the undersigned, declare that I submitted the thesis entitled:

EXPLORING THE STRUCTURAL, MECHANICAL, ELECTRONIC, MAGNETIC, OPTICAL AND THERMOELECTRIC PROPERTIES OF QUATERNARY HEUSLER ALLOYS FeMnScGa, FeMnScAl AND FeMnCrGe

I declare that the work provided in this Dissertation, unless otherwise referenced, is the researcher's own work, and has not been submitted elsewhere for any other degree or qualification.

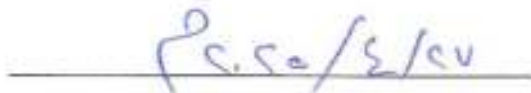
Student's Name:



Signature:



Date:



List of Contents

Dedication	iv
Acknowledgement	v
Declaration	vi
List of Contents.....	vii
List of Tables	x
List of Figures	xi
List of Appendices	xii
Abstract	xv
Chapter One: Introduction.....	1
1.1 Theoretical Framework.....	2
1.2 Spintronics: The Interplay of Charge and Spin for Advanced Electronics	4
1.3 Research Problem and Motivation	5
1.4 Foundations of Material Properties	6
1.4.1 Structural Optimization and Property Prediction.....	6
1.4.2 The Birch-Murnaghan Equation of State	7
1.4.3 Murnaghan's equation of state.....	8
1.4.4 Formation Energy and Material Stability	9
1.4.5 Elasticity and Mechanical Stability.....	11
1.4.6 Electronic Band Structure and Properties of Materials	16
1.4.7 Magnetic Behavior and Spin Configurations	20
1.4.8 Optical Transitions and Spectral Properties.....	22
1.4.9 Thermoelectric Effects and Conductivity.....	26
Chapter Two: Theoretical Frameworks and Advanced Computational Techniques.....	31
2.1 Introduction.....	31
2.2 The Many-Body Problem	32
2.3 Born-Oppenheimer Approximation.....	32
2.4 The variational principle	33
2.5 Hartree-Fock Theory.....	33
2.6 Density Functional Theory	34

2.7 Exchange Correlation Energy Approximation	37
2.7.1 Local Density Approximation	38
2.7.2 Generalized Gradient Approximation	38
2.7.3 The Modified Becke-Johnson Potential.....	39
2.7.4 Augmented Plane Wave (APW) method	39
2.7.5 The Linearized Augmented Plane Wave Method	41
2.8 The WIEN2k code	42
2.9 BoltzTraP Code: A Computational Tool for Transport Properties	43
2.10 The Slack Model for Lattice Thermal Conductivity.....	43
2.11 Computational Methodology	45
Chapter Three: Result and discussion	47
3.1 FeMnCrGe Compound	47
3.1.1 Formation Energy and Structural Properties	47
3.1.2 Elastic Properties.....	50
3.1.3 Phonon Dispersion Analysis	52
3.1.4 Electronic Properties	53
3.1.5 Magnetic Properties and State Stability.....	58
3.1.6 Optical properties	60
3.1.7 Thermoelectric Properties	68
3.2 FeMnScAl Compound.....	73
3.2.1 Structural and Thermodynamic Stability of FeMnScAl	73
3.2.2 Phonon Analysis of FeMnScAl's Dynamical Stability.....	75
3.2.3 Charge Density and Bond Analysis.....	75
3.2.4 Elastic Properties.....	76
3.2.5 Electronic Properties	77
3.2.6 Magnetic Properties	78
3.2.7 Optical properties	80
3.2.8 Thermoelectric Properties	85
3.3 FeMnScGa Compound	88
3.3.1 Structural Properties and Thermodynamic Stability.....	88

3.3.2 Phonon Analysis of FeMnScGa's Dynamical Stability	90
3.3.3 Elastic Properties.....	90
3.3.4 Electronic Properties	91
3.3.5 Electronic Charge Density and Chemical Bonding	92
3.3.6 Magnetic Properties	92
3.3.7 Optical properties	93
3.3.8 Thermoelectric Properties	98
Chapter Four: Conclusions and Future Perspectives	103
4.1 Structural Properties and Stability Analysis	103
4.2 Electronic Structure and Magnetic Properties.....	104
4.3 Optical Properties and Light-Matter Interactions.....	105
4.4 Thermoelectric Properties and Energy Conversion	105
4.5 Technological Implications and Applications.....	106
4.6 Scientific Impact and Methodological Contributions.....	107
4.7 Current Limitations and Technical Challenges	107
4.8 Future Research Directions and Recommendations	108
4.8.1 Experimental Studies	108
4.8.2 Computational Studies	109
4.9 Concluding Remarks and Future Outlook	110
List of Abbreviations	111
References	113
Appendices	127
المُلخَص	ب

List of Tables

Table 1: The optimized lattice parameter a (Å), bulk modulus B (GPa), and its pressure derivative B' (GPa) for FeMnCrGe	50
Table 2: Elastic Properties of FeCrMnGe Quaternary Heusler	52
Table 3: Magnetic Moments of FeMnCrGe Alloy Using GGA and mBJ Methods.....	59
Table 4: Key parameters of ferrimagnetic, paramagnetic, and antiferromagnetic for the FeMnCrGe alloy	60
Table 5: Atomic configuration and structural properties of FeMnScAl.	74
Table 6: Mechanical Properties of FeMnScAl Alloy Configurations at zero strain ($\delta = 0$)	76
Table 7: Magnetic properties of FeMnScAl alloy: Type 1, Type 2, and Type 3 Structures	79
Table 8: Magnetic Phase-Dependent Properties of FeMnScAl (AFM, FM, and PM)...	80
Table 9: Material characterization matrix: spin states and methods	85
Table 10: Spin-up thermoelectric properties at varying temperatures	86

List of Figures

Figure 1: The iterative framework: achieving convergence in Kohn-Sham DFT	37
Figure 2: Schematic representation of the APW	40
Figure 3: Volume optimization results for FeMnCrGe configurations.	49
Figure 4: Three Crystal Structures of FeMnCrGe Compound	50
Figure 5: FeMnCrGe's phonon dispersion curve along high-symmetry paths in the Brillouin Zone	53
Figure 6: Total and Partial Density of States (DOS) for FeMnCrGe Using GGA Approximation.....	54
Figure 7: Total and Partial Density of States (DOS) for FeMnCrGe Using GGA Approximation	55
Figure 8: Band structures of FeMnCrGe: spin-up and spin-down (GGA and mBJ)	57
Figure 9: Two-Dimensional Charge Density Contours of FeMnCrGe in the (110) Plane for Majority and Minority Spins	58
Figure 10: Total Energy Calculations for Different Magnetic Orderings (FM, PM, AFM)	60

List of Appendices

Appendix A: Figures	127
Figure A.1: (a-d) Dielectric function of FeMnCrGe using mBJ and GGA methods	127
Figure A.2: Spin-dependent analysis of refractive index and extinction coefficient with GGA and mBJ.....	128
Figure A.3: Absorption coefficient of FeMnCrGe across spin channels using GGA and mBJ approximations	128
Figure A.4: Optical conductivity of FeMnCrGe in spin configurations using GGA and mBJ	129
Figure A.5: Reflectivity spectrum of FeMnCrGe for Spin channels using GGA and mBJ Methods.....	129
Figure A.6: Energy Loss function of FeMnCrGe in spin configurations using GGA and mBJ	130
Figure A.7: (a-h) Temperature-dependent thermoelectric properties of FeMnCrGe for spin-up and spin-down channels (GGA vs. mBJ)	131
Figure A.8: (a) & (b) Thermoelectric Figure of Merit (ZT) as a function of temperature for FeMnCrGe (GGA vs. mBJ Methods).....	132
Figure A.9: (a) & (b) Temperature dependence of Power Factor (PF) for FeMnCrGe (GGA vs. mBJ Methods).....	132
Figure A.10: (a-b) Hall Coefficient (RH) variation with temperature in FeMnCrGe (GGA vs. mBJ Methods).....	133
Figure A.11: Temperature-Dependent Variation of Lattice Thermal Conductivity in FeMnCrGe Based on the Slack Model.....	133
Figure A.12: Temperature Dependence of Total Thermal Conductivity in FeMnCrGe for Majority and Minority Spin Channels.....	134
Figure A.13: The total energy as a function of unit cell volume for three FeMnScAl configurations is presented.....	134
Figure A.14: The three FeMnScAl crystal structures along with their Brillouin zone and high-symmetry points	135
Figure A.15: The phonon dispersion curve for FeMnScAl along the high-symmetry paths in the Brillouin zone	135
Figure A.16: (a) and (b) Contour plots of charge density for the FeMnScAl alloy	136
Figure A.17: (a-e) Density of states (DOS) of FeMnScAl with total and atomic contributions, computed using the GGA approximation	137
Figure A.18: (a-e) Density of states (DOS) of FeMnScAl, including total and atomic contributions, computed with the mBJ approximation	138
Figure A.19: (a-d) Electronic band structures of FeMnScAl with spin polarization, computed using GGA and mBJ	139

Figure A.20: Energy vs. Magnetic Configuration for FeMnScAl (AFM, FM, PM)	140
Figure A.21: (a) and (b) FeMnScAl's dielectric function determined using GGA and mBJ methods	140
Figure A.22: (a) The refractive index as a function of energy and (b) the extinction coefficient of FeMnScAl calculated using mBJ and GGA methods. ..	141
Figure A.23: The spectrum of absorption coefficient $A(\omega)$ for FeMnScAl alloy, calculated with mBJ and GGA methods	141
Figure A.24: (a) and (b) The FeMnScAl alloy's $\sigma(\omega)$ optical conductivity spectrum, assessed using mBJ and GGA techniques.....	142
Figure A.25: The reflectivity spectrum of FeMnScAl for different spin channels, obtained through GGA and mBJ approaches.....	142
Figure A.26: The energy loss function $L(\omega)$ spectrum of FeMnScAl alloy, studied with mBJ and GGA approaches	143
Figure A.27: (a-h) Thermoelectric Characteristics for Spin-Up and Spin-Down Channels of FeMnScAl Alloy (GGA and mBJ)	144
Figure A.28: Thermoelectric performance metrics (ZT and PF) for spin-up and spin-down channels (GGA and mBJ)	145
Figure A.29: Hall Coefficient (RH) of FeMnScAl Alloy for spin-up and Spin-down channels (GGA and mBJ).....	145
Figure A.30: Lattice and total thermal conductivity (κ_l) of FeMnScAl alloy at varying Temperatures (GGA and mBJ)	146
Figure A.31: Three cubic structural configurations, labeled Types 1, 2, and 3, represent the atomic arrangements of FeMnScGa	146
Figure A.32: Total energy variation with unit cell volume for FeMnScGa types ..	147
Figure A.33: Phonon Spectral Distribution of FeMnScGa	147
Figure A.34: (a-e) GGA-Based Analysis of Total and Atomic DOS Contributions in FeMnScGa	148
Figure A.35: (a-e) mBJ-Based Analysis of Total and Atomic DOS Contributions in FeMnScGa	149
Figure A.36: Spin-polarized electronic band structures of FeMnScGa (a) GGA spin-up, (b) GGA spin-down, (c) mBJ spin-up, (d) mBJ spin-down	150
Figure A.37: 2D charge density contours of FeMnScGa in the (110) plane for (a) majority spin, (b) minority spin	151
Figure A.38: Comparison of Energies and Stability for FM, PM, and AFM Configurations	151
Figure A.39: (a-d) Spectroscopic analysis of the dielectric function of FeMnScGa (0-8 eV)	152

Figure A.40: (a-d) Refractive index and extinction coefficient of FeMnScGa for both spin states	152
Figure A.41: Calculated Absorption Spectra of FeMnScGa (GGA and mBJ) for Both Spin States	153
Figure A.42: Calculated optical conductivity of FeMnScGa (GGA and mBJ) for spin-up and spin-down states	153
Figure A.43: (a) & (b) Reflectivity spectra of FeMnScGa were computed for spin-polarized states using both GGA and mBJ methods	154
Figure A.44: Calculated energy loss function of FeMnScGa (GGA and mBJ) for spin-up and spin-down states	154
Figure A.45: Thermoelectric characteristics of FeMnScGa under varying temperature conditions	155
Figure A.46: Temperature dependence of spin-resolved ZT and PF in thermoelectric performance	156
Figure A.47: (a) & (b) Hall Coefficient (RH) for Spin-Up and Spin-Down Channels in FeMnScGa Using GGA and mBJ	156
Figure A.48: Lattice Thermal Conductivity (κ_l) of FeMnScGa Alloy Across Temperatures Using GGA and mBJ	157
Figure A.49: Total thermal conductivity (κ) of FeMnScGa alloy across temperatures using GGA and mBJ	157
Appendix B: Tables.....	158
Table B. 1: Temperature-dependent ZT and PF values for spin-up and spin-down configurations	158
Table B.2: Hall Coefficient (RH) values for FeMnScAl alloy for specific temperature values	158
Table B.3: Structural and stability parameters of FeMnScGa.....	158
Table B.4: Elastic Constants of FeMnScGa at Zero Strain	159
Table B.5: Partial and Total Magnetic Moments in FeMnScGa Alloy from GGA and mBJ Calculations	159
Table B.6: Total Energy and Structural Parameters for FM, PM, and AFM States for FeMnScGa alloy	159
Table B.7: Key Optical Property Values for FeMnScGa Alloy (GGA and mBJ) ..	160
Table B.8: Thermoelectric Properties of FeMnScGa Alloy	161
Table B.9: K-Point Convergence for FeMnCrGe, FeMnScAl, and FeMnScGa	162
Appendix C: Certificates of acceptance of the research extracted from the dissertation	163

EXPLORING THE STRUCTURAL, MECHANICAL, ELECTRONIC, MAGNETIC, OPTICAL AND THERMOELECTRIC PROPERTIES OF QUATERNARY HEUSLER ALLOYS FeMnScGa, FeMnScAl AND FeMnCrGe

By
Hasan Ali Hasan Masri
Supervisor
Prof. Mohammed Abu-Jafar

Abstract

This dissertation presents a comprehensive analysis of the structural, electronic, magnetic, optical, and thermoelectric properties of three quaternary Heusler compounds: FeMnCrGe (Iron-Manganese-Chromium-Germanium), FeMnScAl (Iron-Manganese-Scandium-Aluminum), and FeMnScGa (Iron-Manganese-Scandium-Gallium), using first-principles theoretical frameworks. This research relies on the density functional theory (DFT). The calculations use the Perdew–Burke–Ernzerhof (GGA-PBE) functional. This approach uses a generalized gradient approximation. It ensures precise electronic structure analysis. Additionally, it uses the modified Becke-Johnson (mBJ) functional. These approaches help to evaluate the material's potential applications in spintronics and energy conversion devices. All three compounds have a face-centered cubic structure with adherence to the Fm-3m space group. A negative formation energy characterizes their stable configurations. This confirms their stability. Moreover, they comply with mechanical stability conditions.

FeMnCrGe (Type 1 structure), FeMnScAl, and FeMnScGa (Type 2 structures) all meet mechanical stability requirements and exhibit structural stability through a hybrid ionic-covalent bonding framework. The electronic structure analysis depicts strong half-metallic behavior for all compounds studied, with 100% spin polarization. The FeMnCrGe compound has a spin-down bandgap value calculated as 0.974 eV with a modified Becke-Johnson potential, compared with a 0.1574 eV bandgap for FeMnScAl and 0.442 eV for FeMnScGa. As per the Slater-Pauling rule, magnetic moments have been calculated. The FeMnCrGe compound illustrates a magnetic moment value per formula unit as 1.00 μ_B , with FeMnScAl and FeMnScGa having magnetic moments of 3.00 μ_B each. Such a fluctuation is attributed to strong ferromagnetic behavior, which is substantiated through strong magnetic interactions from manganese. Investigation of the

thermoelectric properties reveals high efficiency in energy conversion. The material FeMnScAl has a peak value for ZT of 0.689 at 150 K, and FeMnCrGe has a value of 0.42 at 500 K.

Also, the optical properties of all three compounds demonstrate high ultraviolet absorption, with FeMnScGa having absorption coefficients above $180 \times 10^4/\text{cm}$. Additionally, all materials exhibit a high refractive index, and large interband transitions are present. Heusler compounds are multifunctional. They show promise in spintronics, thermoelectrics, and optoelectronics. These applications are relevant to electronics and energy harvesting.

Keywords: Quaternary-Heusler alloy; electronic structure; magnetic properties; Half-metallic; elastic; optical; thermoelectric; FP-LAPW.

Chapter One

Introduction

Meeting the energy and technological demands of the planet requires the discovery of new materials with functionally tailored properties. With industries working on tapping into renewable power and developing future electronic devices, researchers explore materials that can efficiently convert, store, and handle energy. The future uses of energy harvesting with green energy, high-frequency computation, data storage, and quantum technologies demand materials with higher function. To meet such needs, it is critical that materials have tunable electronics, strong magnetism, and ideal thermal conductivity. Heusler compounds have been considered a very promising class of materials, exhibiting excellent flexibility and exceptional properties. They were first discovered well over a century earlier, in 1903. Heusler compounds have continued to be at the center of condensed matter physics and materials science. Heusler compounds have a unique structure at the molecular level, complemented by a wide range of compositions, thus allowing fine-tuning to various properties. They have a wide range of electrical, magnetic, optical, and thermoelectric properties, thus being useful in the context of green energy technologies, data storage improvement, and the application of the field of spintronics, which uses electron spin to process information. Based on the nature of the alloys' specific composition and structure, it is possible to precisely engineer the materials to be used as semiconductors, metals, or half-metals. Traditional Heusler compounds fall under two major classes: full Heusler alloys, as represented by the formula X_2YZ , and half Heusler alloys, by the formula XYZ . In both classes, X and Y tend to be rare or transition metals, and Z is a main group element. Developments in materials science in recent years led to the discovery of the quaternary Heusler alloys, which represent a subclass that maximizes the function and potential uses of the compounds. Quaternary Heusler alloys have a 1:1:1:1 stoichiometry and $XX'YZ$ structure, thus allowing other transition metals to be added, which in turn allows new scope to tailor their properties. This property has opened the door to tailoring electronic, magnetic, and thermoelectric properties to exactly meet the needs of new technologies. The quaternary Heusler alloys are especially noteworthy due to their high spin polarization, unusual optical properties, and excellent thermoelectric performance. Their potential applications include spintronics, thermoelectric systems, and optoelectronic

devices, which makes them quite promising. Still, numerous quaternary Heusler alloys have not been adequately studied, particularly concerning the influence of atomic arrangements and compositions on their multifunctionality. In this study, we focus on FeCrMnGe, FeMnScAl, and FeMnScGa a quaternary Heusler alloys with promising multifunctional properties. Using computational modeling and DFT calculations, we investigate structural, electronic, magnetic, optical, and thermoelectric properties. The research work will join the available literature on Heusler alloys and provides the basic information which may be helpful in designing new materials in view of technological development belonging to the next generation.

1.1 Theoretical Framework

Quaternary Heusler alloys (QAH) are extensively studied in materials science because of their outstanding electronic, magnetic, and mechanical characteristics. Quaternary Heusler alloys (QAH) exhibit remarkable properties, making them highly promising candidates for applications in spintronics, magnetoelectronics, and thermoelectric devices [1,2]. The following literature review will discuss the major applications and research results of recent studies on quaternary Heusler alloys, thus highlighting their importance in high-technology applications.

Heusler alloys are of specific relevance for applications in spintronics and magnetoelectronics, where high spin polarization and half-metallicity are critical requirements for information storage and processing in spin-transfer torque (STT) memory cells and magnetic tunnel junctions (MTJs) [3,4]. High spin polarization and high Curie temperatures are observed in alloys such as CoRhMnZ and CoFeMnSi, which are therefore excellent choices for applications in spintronic devices that are operable at elevated temperatures [5,6]. Compatibility with the lattice of MgO substrates is also attained by altering the alloys, i.e., the replacement of Rh with Co, which enhances the spin-transfer efficiency of STT devices [7]. Quaternary Heusler alloys (QHAs), especially those composed of 3d and 4d transition metals, have inspired interest due to their intriguing thermoelectric, optical, and spintronic properties, as demonstrated by the emergence of alloys such as FeCrTiSb [8], FeCrRuZ (Z = Al, Ga, In, or Si) [9], and YFeCrZ (Z = Al, Sb, or Sn) [10].

The significant half-metallicity and spin polarization exhibited by NiFeCrX (X = Al, Si, Ge, In), AgCoFeX (X = Al, Ga, Si, Ge, Sn), and NdCoMnGa, strongly highlight their suitability for spintronics applications [11].

CoFeTiSn was identified to show a stable spin-polarized phase, thus proving its applicability in spintronic devices and future memory technology, highlighting the relevance of Heusler alloys [12]. Additionally, high spin polarization and low Gilbert damping (a parameter that quantifies the dissipation of energy during magnetization dynamics, affecting how quickly magnetic moments stabilize after perturbation) are characteristic of alloys like CoFeXGe (where X is Hf or Ta) and CrTiCoZ (with Z as Al, Ga, or In) [13,14].

Beyond spintronics, many quaternary Heusler compounds are also under investigation for their potential in solar energy conversion. Featuring strong absorption of visible light, appropriate electronic energy gaps from 0.30 to 2.50 eV, and excellent dielectric screening, compounds like NiTiSn, PdLiScPb, PdLiYPb, PtLiScGe, and CoLiHfGe indicate exceptional potential for use in photovoltaic and thermoelectric applications [15]. Similarly, (CoLiZrSn, PtLiYSn, NiLiZrGa, FeLiVSi, PdLiYPb, CoLiTaAl, NiLiZrAl, FeLiNbGe, NiTiSn, CoLiNbAl, PtLiScGe, PdLiYSn, CoLiNbGa, CoLiHfGe, PdLiScPb, PtLiYSn, and CoLiHfSn) have emerged as a standout material for spintronics, thermoelectric, and optoelectronic uses due to their stable ferromagnetic ground state, full spin polarization, and high ZT value [15].

In addition to their other qualities, thermoelectric properties are crucial for Heusler alloys. Many of these compounds demonstrate significant relationships among thermal conductivity, electronic structure, and resistivity, which vary with temperature, rendering them appropriate for thermoelectric devices. The unique resistance anomalies occurring at the Curie points of compounds like CoRuMnGe and CoRuVZ where Z is Al or Ga put them at the center stage for applications for thermoelectric cooling and thermoelectric power generation [16]. Extremely high values of the figure of merit (ZT), determined in the case of LiHfPdX where X is Al, Ga, or In, along with that of FeZrCrX where X is Si, Ge, or Sn, also prove their feasibility for thermoelectric applications [17,18].

Besides their magnetic and electronic properties, the mechanical stability and high Curie temperatures of quaternary Heusler alloys make them reliable in demanding environments. Exhibiting high bulk moduli and strong resistance to structural deformation, alloys in the ZrFeVZ family ($Z = \text{Al, Ga, In}$) maintain reliable performance even at elevated temperatures [19]. Given their properties, these alloys are well-suited for demanding applications, including automotive sensors and aerospace parts, where they must withstand high thermal and mechanical stress [20,21].

The combination of tunable magnetic, electronic, and thermal characteristics in these materials makes them highly suitable for use in spintronic and thermoelectric technologies. With increasing knowledge of alloy behavior under diverse conditions, quaternary Heusler compounds are projected to gain prominence in the development of high-efficiency and energy-efficient electronic systems across different industries.

1.2 Spintronics: The Interplay of Charge and Spin for Advanced Electronics

Spintronics, commonly referred to as spin electronics, is an innovative field that combines the electron's spin with its charge for technological applications. By contrast, standard electronics limits itself to manipulating charges, whereas spintronics uses spin effects to enhance performance in communication and data technologies significantly. The concept became widely recognized after Peter Grünberg and Albert Fert's pioneering work on giant magnetoresistance (GMR) in 1988, which revolutionized hard drive technology and inspired detailed investigations into spin-dependent transport phenomena [22].

Electron spin a quantum mechanical degree of freedom manifesting as bistable "up/down" polarization states forms the operational basis of spintronic systems. In ferromagnetic materials, the splitting of majority and minority spin bands at the Fermi energy level results in spin-polarized conduction, a phenomenon first described by Mott in 1936 and further developed in the two-current model by Fert and Campbell in the late 1960s. This spin-polarized conduction forms the basis for spintronic devices.

Giant Magnetoresistance (GMR) is a phenomenon in which the electrical resistance of magnetic multilayers varies markedly depending on the relative orientation of their magnetic moments being minimal when the moments are aligned parallel and maximal

when aligned antiparallel. This effect has been applied in hard disk drive read heads, allowing for substantial increases in storage densities [23].

A crucial effect in magnetic tunnel junctions (MTJs) is Tunneling Magnetoresistance (TMR), in which spin impacts electron tunneling across an insulating barrier. The effect has significantly contributed to the progress of MRAM technology by enabling stable nonvolatile memory with fast read and write operations [24].

Magnetic state engineering via Spin-Transfer Torque (STT) bypasses external field reliance through spin-polarized current injection, a paradigm-shifting approach critical to developing power-efficient MRAM architectures and high-frequency spin-torque oscillators [25].

1.3 Research Problem and Motivation

The remarkable mix of high spin polarization, outstanding optical and thermoelectric properties, along with superior half-metallic ferromagnetism in quaternary Heusler alloys, has drawn considerable interest. As a result, they are seen as promising candidates for future spintronics and energy harvesting applications [16]. This study aims to investigate the exceptional properties of quaternary Heusler alloys through the structural optimization and comprehensive characterization of newly designed compounds: FeCrMnGe, FeMnScAl, and FeMnScGa. We apply state-of-the-art first-principles calculations based on the well-tested Wien2K method in order to perform this complete investigation, thus enabling precise calculations of material properties and electronic structures.

FeCrMnGe is differentiated by its novel combination of half-metallicity and high spin polarization, both of which possess great significance for the improvement of high-efficiency spintronics. Despite its promising characteristics, no experimental or theoretical data currently exist regarding its properties. To address this gap, our research will systematically explore the elastic, electronic, magnetic, optical, and thermoelectric properties of FeCrMnGe. Explanation of FeCrMnGe properties is very crucial in determining its prospects for application in real-life electronic devices and energy conversion. Furthermore, its alloy system, FeMnScAl, is relatively less studied in the current literature, especially in relation to its application prospects in areas involving thermoelectricity and optics. The major aim in this research is to undertake an extensive

study on FeMnScAl based on its structural properties, electronic properties, magnetic properties, optical properties, and thermoelectric properties.

This work is intended to present fresh perspectives on FeMnScAl's versatility and multifunctionality for further increasing its future applications in green energy technologies and in the field of optoelectronics. This work also examines the as yet unknown properties of FeMnScGa and specifically its mechanical properties as well as its optical properties. Utilizing its unique approach, the research performs an intensive investigation of three unique structural arrangements to determine how stable each layout is.

Furthermore, the optical characteristics of FeMnScGa will be analyzed, specifically for their potential applications in advanced green energy technologies, where both high performance and stability are essential. This methodology boosts our predictive power. We can better understand the properties of FeCrMnGe and FeMnScGa. Moreover, it provides a solid base. These base supports research into other promising quaternary Heusler alloys. These alloys have potential in green technologies. These alloys have implications for emerging green technologies. This research sets a foundation. The foundation is for future studies on Heusler alloys. It also aids in designing new materials.

1.4 Foundations of Material Properties

1.4.1 Structural Optimization and Property Prediction

Structural properties, including bonding and the molecular and atomic arrangement at the atomic level, govern the behavior of materials. A material's structure directly affects properties such as conductivity and strength. Unit cell size determines material properties. Atomic arrangement also plays a role. Equations of state (EOS) link these structural properties to external factors. These factors include pressure and temperature. EOS helps predict material behavior under various conditions. Moreover, equations of state (EOS) correlate structure parameters with external conditions, i.e., pressure and temperature, to predict material responses in different situations.

1.4.2 The Birch-Murnaghan Equation of State

The Birch-Murnaghan Equation of State (EOS) is a mathematical expression in the field of materials science that explains the interdependence of bulk modulus, pressure, and volume, especially in situations involving finite strain. Materials science uses inverse proportionality. It explains the relationship between pressure and volume. Increased pressure typically reduces volume. Conversely, decreased pressure increases volume. The equation of state is of major importance to both condensed matter physics and geophysics since it is used to predict the behavior of material under the action of high pressure and hence offers a lot of insight into the compressive and elastic behavior of materials. The equation of state is named after the names of both Francis Birch and Francis Dominic Murnaghan since both researchers made major contributions towards the theoretical background on which this theory is based. Birch applied Murnaghan's theoretical framework to explain the behavior of materials under geological pressures, while Murnaghan created the strain-energy function approach that formed the basis for this model. The Birch-Murnaghan EOS can be expressed in its third-order form as [26]:

$$E(V) = E_0 + \frac{9V_0 B_0}{16} \left\{ \left[\left(\frac{V_0}{V} \right)^{\frac{2}{3}} - 1 \right]^3 B_0 + \left[\left(\frac{V_0}{V} \right)^{\frac{2}{3}} - 1 \right]^2 \left[6 - 4 \left(\frac{V_0}{V} \right)^{\frac{2}{3}} \right] \right\} \dots \dots \dots (1)$$

In this relationship, V denotes the volume occupied by the material, with V_0 representing the equilibrium volume under zero pressure. The Birch-Murnaghan equation of state (EOS) represents a pressure-volume relationship and enables an expression for energy as a function of volume. Understanding a material's structure is dependent on conclusions reached through a relationship of this nature. This formalism explains interdependency among pressure, energy, and volume. Additionally, it gives good insights into material stability. The compression number, symbol B , is one of the parameters defining Birch-Murnaghan EOS and determines how the material opposes pure bulk compression.

The EOS measures a material's resistance to bulk compression quantitatively. Also, the resistance to compression for a material is an intrinsic property of its stability and, by extension, its suitability for use at high pressures. A large bulk modulus indicates high resistance to compression. Conversely, a small bulk modulus indicates the material is easily compressed. This property is key to figuring out how materials hold up under

high pressures, as it influences both their initial resistance to compression and how that resistance evolves when strain is applied. With applications in multiple research disciplines, the Birch-Murnaghan EOS is particularly valuable in high-pressure physics for studying strain effects, material behavior under extreme conditions, and structural stability in aerospace and defense technologies. There are many extensions and modifications or alterations, specifically of the Birch-Murnaghan EOS, as well as the town of fourth-order in addition to first- and second-order Birch-Murnaghan pieces of information, with higher-order use of adding terms and shown specifically to the energy-pressure volume, specifically to experimental conditions of pressure range. Alternative models like the Murnaghan EOS provide different approaches for specific materials and scenarios, expanding the applicability of pressure-volume relationships in a broader range of contexts.

1.4.3 Murnaghan's equation of state

Pressure, temperature, and density are interconnected in condensed matter. The MEOS is a key model for analyzing these relationships. It investigates the links between pressure, temperature, and density. Despite the availability of several forms of the EOS, Murnaghan's remains one of the earliest and most frequently applied. Under finite strain, the model presumes that there is a linear dependence between a solid's bulk modulus and pressure [27,28].

$$P(V) = \left(\frac{B_0}{B'_0}\right) \left[\left(\frac{v_0}{V}\right)^{B'_0} - 1 \right] \dots\dots\dots(2)$$

In this context, P is the applied pressure, V is the volume of the material in response, V_0 represents the equilibrium volume, B_0 is the bulk modulus at zero pressure, and B'_0 quantifies how the bulk modulus varies with pressure when at zero pressure. Ground-state properties of materials are computed using DFT at 0 K to avoid the effects of Fermi-Dirac occupation probability, with zero Pascal (Pa) conditions applied to exclude external influences. The electronic ground state energy $E(V)$ can be obtained by integrating the pressure equation, since $P(V) = -(\frac{dE(V)}{dV})$. Two key formulations for the energy-volume relationship have been proposed in the literature: The first, obtained by direct integration, is as follows:

$$E_a(V) = \frac{B_0 V}{B'_0} \left(\left(\frac{V_0}{V} \right)^{B'_0} - 1 \right) + C_a \dots\dots\dots (3)$$

The second formulation modifies the integration constant and is represented as:

$$E_b(V) = \frac{B_0 V}{B'_0(B'_0-1)} \left(\left(\frac{V_0}{V} \right)^{B'_0} - 1 \right) + C_b \dots\dots\dots (4)$$

While these forms are mathematically equivalent, they behave differently when used in least-squares fitting procedures. The second version E_b is chosen because it delivers genuinely independent fitting parameters and greater numerical stability during the fitting procedure. This is because it has a well-defined minimum at $V = V_0$ and its least-squares fitting function shows a rectilinear valley in parameter space, making it more suitable for numerical optimization.

1.4.4 Formation Energy and Material Stability

A critical thermodynamic quantity, formation energy (E_{Form}), is essential for understanding the stability and chemical reactivity of materials. This principle elucidates the conditions under which compounds form spontaneously and remain stable, making it indispensable in materials design and chemical synthesis. The formation energy, in a more general representation, refers to the energy change assisted by the process of compound formation from constituent elements in their standard. E_{Form} is the main thermodynamic parameter that reflects the energy difference between a compound and its base elements in their standard state, telling much about both stability and synthesizing easiness, as illustrated in the equations below [29,30]:

$$E_{Form} = E_{compound} - \sum_{i=1}^n E_{element(n)} \dots\dots\dots (5)$$

This equation encapsulates several key components.

The energy of a compound is represented as $E_{compound}$, and $E_{Elements(n)}$ represents the energy of the elements in their stable form. The coefficients (n) represent stoichiometric ratios of each element included in a compound. The coefficients (n) correspond to the stoichiometric ratios of the elements in the compound. This value measures compound stability at standard conditions (0 K, 0 atm).

The sign and magnitude of the formation energy carry significant meaning. A negative formation energy indicates an exothermic process, meaning energy is released during compound formation. The negative formation energies reflect the stability of the compounds and their ease of formation, whereas the positive formation energies indicate endothermic processes and thereby require energy for the formation of the compounds. The more negative the formation energy, the greater the thermodynamic driving force for compound formation and the higher its stability relative to its constituent elements [31].

Advancements in computational methods, particularly Density Functional Theory (DFT), have greatly enhanced the precision and complexity of formation energy calculations in modern materials science. Theoretical approaches predict formation energies of hypothetical compounds, accelerating materials discovery. This predictive capability has driven significant advancements in areas such as catalysis and battery technology, showcasing the extensive utility of formation energy. In catalyst design, the formation energy gives insight into the stability of catalytic materials under reaction conditions. In semiconductor studies, it assists in new electronic material development by pointing out which dopant configurations will most probably be stable. In energy storage applications, formation energy calculations help identify promising electrode materials for batteries and fuel cells [32].

This principle is of particular importance in phase transformation and material degradation research. The researchers study the energy of phase formation with respect to different phases and states to improve predictions about material performance so as to maintain reliability and stability under various conditions. Research in this area is very important in applications requiring structural stability under harsh conditions, including high-temperature and corrosive conditions.

The formation energy as an important descriptor has promoted improved efficiency in computational material databases used for the screening of new materials. Researchers have been able to speed up screening in databases based on formation energy and calculated properties for compounds and thus select ideal candidates for intended applications. This methodological approach has significantly improved material discovery as well as material optimization processes.

Formation energy is an important source of information in the design of chemical reactions. Chemists use this information to predict reaction products, develop new synthesis pathways, and understand how to control conditions, including the suppression of unwanted side reactions or decompositions [33].

Advances in computational capacity and experimental technique over recent times have dramatically altered the study of formation energy. Modern approaches now utilize artificial intelligence and machine learning based on proven thermodynamic principles to yield very accurate formation energy predictions [34]. These combined approaches now hold promise for material design and material optimization that could lead to clean energy breakthroughs, innovations in electronics, and applications in the field of green chemistry.

1.4.5 Elasticity and Mechanical Stability

The way a material deforms under pressure and subsequently recovers its shape is determined by its elastic properties. Elastic constants quantify these properties, defined by particular parameters that connect stress with strain across various directions. Through these constants, critical information about a material's deformation resistance can be obtained, indicating whether it responds with elastic, plastic, or brittle properties when external forces are applied. The stiffness matrix C describes the elastic response through the second derivative of the energy E with respect to strain ϵ [35].

$$C_{ij} = \frac{1}{V_0} \frac{\partial^2 E}{\partial \epsilon_i \partial \epsilon_j} \dots\dots\dots (6)$$

Where V_0 is the equilibrium volume of the crystal. Stability in elastic materials is verified when the matrix C has positive eigenvalues or, equivalently, when each leading principal minor is positive, as per Sylvester's criterion. For cubic crystal systems, the elastic stability conditions are derived from the fact that the cubic symmetry greatly simplifies the form of the stiffness matrix C , leaving only three independent elastic constants: C_{11} , C_{12} , and C_{44} . The cubic system's stiffness matrix in Voigt notation appears as [35]:

$$C_{\text{cubic}} = \begin{bmatrix} C_{11} & C_{12} & C_{12} & 0 & 0 & 0 \\ C_{12} & C_{11} & C_{12} & 0 & 0 & 0 \\ C_{12} & C_{12} & C_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{44} \end{bmatrix} \dots\dots\dots (7)$$

The necessary and sufficient conditions for elastic stability in cubic crystals are known as the Born stability criteria. These criteria ensure that the matrix C_{cubic} is positive definite, which corresponds to the crystal being mechanically stable under small deformations. For a cubic crystal to be mechanically stable under small deformations, its stiffness matrix C_{cubic} must be positive definite, as ensured by these criteria. The following inequalities define the stability conditions for cubic crystals: $C_{11} > 0$, $C_{11} + 2C_{12} > 0$, $C_{11} - C_{12} > 0$, and $C_{44} > 0$ [35]. Four conditions ensure C (cubic) is positive definiteness for stability against small mechanical perturbations. Under standard DFT conditions, elastic constants (C_{ij}) were calculated to assess the mechanical stability of quaternary Heusler alloys. Using the Reuss approximation, the bulk modulus (B), shear modulus (S_R), Young's modulus (Y), Poisson's ratio (ν), and anisotropic factor (A) were determined [36–39].

Bulk modulus (B), which measures the resistance of a material to change in volume under hydrostatic pressure, is a critical property. The modulus describes the degree to which the material can sustain deformation, including compression and expansion. The bulk modulus also measures the degree of volume change in a material due to hydrostatic pressure. Expressed mathematically as $B = -V \frac{dP}{dV}$, the bulk modulus is defined as the ratio of a small increase in pressure to the relative decrease in volume [40]. A high bulk modulus signifies resistance to compression; thus, materials with this property require substantial pressure for volume changes, enabling faster sound wave propagation. Such resistance to alterations in volume guarantees stability in environments where high pressure is present. In fluid mechanics, this modulus quantifies gas and liquid compressibility, and its experimental measurement involves techniques like hydrostatic compression, nanoindentation, or ultrasound [41]. Theoretical predictions are also possible using density functional theory (DFT), which considers atomic interactions. It can be derived from the elastic constants, as defined by the Reuss relation:

$$B_R = \frac{1}{3}(C_{11} + 2C_{12}). \dots\dots\dots (8)$$

Also known as modulus of rigidity, shear modulus is among the important material properties that measure the response of a material to shear stress. The shear modulus is the ratio of shear stress to shear strain in the linear elastic range of the material's stress-strain curve. The shear modulus, S_R , on the other hand, is simply shear stresses τ (force per unit area) acting on shearing strains δ (the deformations caused by the application of shear stresses) and is written mathematically as $S_R = \frac{\tau}{\delta}$. The shear modulus (S_R) is again defined as the measure of a material's resistance to shear, a force that wants to make layers of the material slide over each other. Shear modulus is useful for predicting how any given material will react to torsion or shearing loads, and it represents stiffness, elasticity, and shear strength, which all go a long way in beam, shaft, and building design. It also helps evaluate responses to pressure and tension. Each of the standard measurements (torsion testing, shear wave ultrasound, and nanoindentation) should be accompanied by an equipment reaction [42]. They can also be predicted via computational methods, such as molecular dynamics, finite element, and density functional theory, which study atomic structures and interactions of the material.

Elastic constants serve as input for calculating it using the Reuss relation:

$$S_R = \frac{5C_{44}(C_{11}-C_{12})}{4C_{44}+3(C_{11}-C_{12})}. \dots\dots\dots (9)$$

The bulk modulus (B) and shear modulus (S_R) material properties are analyzed in the B/S_R ratio to characterize material properties and provide information about several mechanical properties. Bulk modulus indicates the capacity of a material to respond to uniform compressive pressure changes in volume. The shear modulus defines the capacity of a material to react to shape changes when it is subjected to a shear load. Therefore, the B/S_R ratio sums up these properties and is a criterion for ductility. Generally, the greater the B/S_R ratio of a material, the more ductile it is, while low-ratio metals are usually brittle and fracture under low-deformation stresses.

Materials scientists use the B/S_R ratio to categorize materials as either ductile or brittle. This classification is key to designing structural components. In geology, it plays a role in seismic analysis. It forecasts how geologic formations behave under stress, helping to

assess earthquake risks. In the engineering design of buildings, knowing the significance of the B/S_R ratio is important to determine if a building can support static and dynamic forces, such as loads acting on a structure amidst an earthquake or strong winds. The B/S_R ratio is about material stiffness; eye glancing at higher values essentially means greater resistance to compression/expansion and a lower “instability.”

This ratio is important for designing earthquake-resistant structures, helping to choose materials that effectively balance flexibility and strength to endure seismic stress. The B/S_R ratio in anisotropic materials indicates that the property changed with respect to direction. This aspect is especially crucial in composite structures. Typically a small B/S_R ratio would indicate that the material is ductile, while a larger ratio would encapsulate a brittle behavior that assists designers selecting materials for mechanical behavior. The B/S_R ratio is a significant parameter in that it assists the designer with values accounting for material design, safety, and performance under stress conditions, leading to better material uses in a variety of fields [43].

Bonding types, such as ionic, covalent, and metallic, can influence Poisson’s ratio ν , though they do not solely determine it due to additional factors like microstructure and porosity. Generalizations based on bonding type are unreliable, as materials with similar bonding can display a wide range of Poisson’s ratios depending on structural and compositional specifics. Poisson ratios below 0.25 suggest ionic bonds, whereas those above 0.25 suggest covalent bonds [44]. Poisson’s ratio can be calculated by taking the negative ratio of lateral strain to axial strain or derived from bulk modulus B and shear modulus S_R using the following equation:

$$\nu = \frac{3BS - 2S_R}{2(3B + S_R)} \dots\dots\dots (10)$$

Young's modulus (Y) is a parameter used to describe the stiffness of a material and provides a measure of the relation between stress and strain during events of uniaxial compression or tension. It is represented as the proportion of stress (force per unit area) to strain (deformation), having the mathematical description Y equals strain over stress [45]. Structural design, material comparison, and mechanical performance evaluation are dependent on Young’s modulus, which predicts flexural deformations for a material under loading conditions. Young’s modulus is affected by several different parameters, including composition, microstructure, temperature, and porosity, all affecting stiffness

[46]. In terms of stiffness, the magnitude of the modulus varies considerably between materials. Metals have high values, representing higher stiffness, while polymers have lower values, representing higher flexibility. The variation is an important factor in material selection for particular applications. Young's modulus is measured by loading and measuring deformation in an experimental sample. Moreover, Young's modulus is identified with the slope in a curve plotted for stress against strain [37]. Moreover, Young's modulus is calculated based on bulk and shear modulus values as follows:

$$Y = \frac{9BS_R}{(S_R+3B)} \dots\dots\dots (11)$$

The anisotropic factor (A) is an indicator of how physical or mechanical properties, i.e., elasticity, differ depending on directions within a material. Anisotropy is a term that relates to direction dependence, meaning that response to applied loads and stresses is dependent on their direction. On the contrary, isotropy is a term that is used to define uniformity throughout all directions, where the properties show equivalent behavior regardless of direction and application of load.

In materials science, the anisotropic factor is used to quantify the degree of directional variability in a material. For cubic materials, the anisotropic factor A is calculated using the material's elastic constants with the relation:

$$A = \frac{2C_{44}}{(C_{11}-C_{12})} \dots\dots\dots (12)$$

Experiments or computational modeling can determine these constants. These constants provide a measurable basis. This basis helps understand a material's directional stiffness. The anisotropic factor indicates how elasticity varies in a material with direction-dependent properties. When the anisotropic factor, A , equals 1, the material is isotropic. Anisotropic behavior arises when a material's properties change with direction, rather than its dimensions. Its elastic properties vary depending on the measurement direction. In contrast, if the anisotropic factor, A , equals 1, the material is considered isotropic. This means its stiffness is the same in all directions [37,47,48].

In composite substances, where the phases are layered or combined to impart strength, anisotropy becomes a pertinent factor. In addition, it affects geophysics because the type of stress that exists in the rock helps to stabilize its layers. Similarly, in design, the

type of force that exists on the loads may require the loads to be positioned in certain manners to ensure structural stability as well as function. The anisotropic factor becomes the central parameter in structural design and selection, playing a critical role in the production of components designed to endure directional stresses. Its applications are unmatched across various fields, including geophysics and emerging engineering disciplines.

1.4.6 Electronic Band Structure and Properties of Materials

The electronic properties determine how a material responds to parameters outside itself and, hence, its electrons. The complex arrangement of electrons in energy bands is instrumental in determining a material's unique electronic properties and their manifestation. The structural arrangement of a material leads to an overarching framework that aids in understanding electronic state distribution in different energy ranges in solid materials. The distribution of energy states highlights major properties, i.e., conductivity, mobility, extent of localization of electrons, and interaction with ions, adjacent electrons, and outside parameters. Essential properties like conductivity, mobility, and degree of confinement have a relationship with each other in this context as well as with the actions of ions, adjacent electrons, and outside parameters. The band structure comes out as a consequence of the atomic structure of a material and interatomic interaction and controls how electrons behave relative to material parameters in a context particularly involving periodic behavior in electronic material in crystals [49]. In solid substances, the complex structure holds electrons in specific energy levels that they are allowed to occupy. Because of the overlap of atomic orbitals caused by densely packed atoms in the structure, these energy levels become extended bands [50]. The energy states arrange themselves into bands that are divided by gaps. In conductors, this setup facilitates electron movement through overlapping bands that permit free flow, while in insulators, wide gaps prevent electron mobility.

At absolute zero, electrons fill the valence band, which represents their highest energy states. The conduction band, positioned above, allows them to absorb enough energy to move without restriction. Compared to insulators and semiconductors with their distinct band gaps, conductors may exhibit overlapping band structures. Electric fields trigger the movement of conduction band electrons, a process fundamental to the conduction of electrical current [51]. The band gap, or the energy difference between the valence and

conduction bands, is fundamental in distinguishing materials. Insulators have large band gaps (>4 eV), making electron transition between bands difficult; semiconductors have moderate gaps (typically 0.1–4 eV), allowing thermally or optically excited electrons to jump the gap; and conductors have no gap or overlapping bands [52]. The band gap size therefore directly determines the material's classification and electronic uses.

Symmetry of the crystal structure, interatomic spacing and material composition all contribute towards defining the material's band structure. Furthermore, pressure, temperature, and magnetic fields from outside also affect the band structure by changing the potential energy profile seen by electrons. Furthermore, changes in composition in the form of alloy and doping also influence electronic states; they are crucial processes used in semiconductor engineering [53].

Experimental methods like Angle-Resolved Photoemission Spectroscopy (ARPES) and X-ray Photoelectron Spectroscopy (XPS) probe band structures with direct electronic state measurements. Additionally, electronic bands are also computationally simulated using approaches like Density Functional Theory (DFT), involving solutions to many-body Schrödinger equations for electrons in periodic settings [54].

The electronic properties of a material show how its electrons behave when subjected to an external stimulus, including electric and magnetic fields and changes in temperature. These reactions depend on how electrons fill energy bands, specifically the valence and conduction bands. The structures of these bands form the basis for understanding how electronic states fill various energy ranges in solid materials. Energy state structure highlights specific properties like conductivity, mobility, electron localization, and coupling with ions, adjacent electrons, and exterior fields. The structure carries with it critical attributes like conductivity, mobility, electronic localization, and coupling with ions, nearby electrons, and exterior surroundings. The structure arises directly out of atomic structure and interatomic interaction and goes on to determine a material's properties and thus its electronic behavior; specifically found during an examination of solids, e.g., crystalline structures, electronic behavior is influenced due to periodicity [49]. Within solid structure, electrons remain confined within specific energy ranges and thus lead to the formation of continuous bands due to orbital overlap among the solid structure's constituent atoms as a consequence of close packing among these constituent atoms [50]. The energy states form an array of bands separated by gaps. For

conductors, overlap among these bands allows for smooth movement by electrons while insulators have broad gaps that resist electron movement. At absolute zero temperature, electrons occupy the valence band, being the highest energy level below the conduction band to allow them to have enough energy to move freely. For conductors, overlap is possible among the band construction compared to distinct gaps found in insulators and semiconductors. Current flow is allowed by electrons being in the conduction band upon application of an electric field [51]. The energy gap is referred to as that energy difference existing among valence and conduction bands and is crucial for material classification into various categories. Insulators have broad gaps (above 4 eV), while semiconductors have intermediate gaps (from 0.1 to 4 eV), allowing electrons to overcome the gap through photonic and/or thermally induced excitation; however, conductors have zero gap or overlapping bands [52]. Therefore, gap width is used as a direct parameter for characterization and decides on electronic application for a material. Configuration and energy for a given band structure is altered by outer parameters that change electrons' environment relative to their energy in terms of potentials. Environmental variables including pressure, temperature, and magnetic fields have the ability to change a given structure for bands by changing the profile for electrons' potential energy. Electronic states are altered by variation in composition including processes such as alloying and doping that form a critical part for semiconductor technology [53]. The structure is studied experimentally using methodologies including Angle-Resolved Photoemission and X-ray Photoelectron Spectroscopy, which act as direct instruments for electronic state determination. In computational calculations towards visualizing electronic bands, computational aids like Density Functional Theory (DFT) are used to solve the many-body Schrödinger equation for electrons in a periodic potential [54]. The idea of a Brillouin zone (BZ) is very important in solid-state physics and is used as the unit cell for the reciprocal space in wavevector (k-space). The nature of reciprocal space shows that only a finite number of wavevectors is able to fully capture the periodicity in a crystal structure; this is an important aspect in defining energy allowed for electrons in a solid. Construction of a Brillouin zone is achieved through drawing perpendicular bisectors from center to each reciprocal space point and thus dividing space into areas closer to one point than to any other point. The resulting polyhedral structure makes up the first Brillouin zone and is

considered to be the smallest and most important zone as it encloses all unique information related to periodic potential and electronic properties.

In a body-centered cubic (BCC) lattice band structure analysis, the high-symmetry path along line Γ -X-W-K- Γ -L-U-W-L within the Brillouin zone is used conventionally. Point Γ (Gamma) represents the center of Brillouin zone ($k = 0$), and X represents an edge center lying along one face area of the zone. Point W marks an edge corner in the Brillouin zone and point K marks an edge center in a region between adjacent faces. Point L is used to mark center of zone face and point U lies on another zone edge. The Brillouin zone is that comprehensively enhances electronic band structure representation with its ability to show wavevector mapping as well as calculate energy eigenvalues that highlight crucial features including conductivity and electronic localization properties. It illustrates how energy and momentum of electrons vary in a crystal and is therefore very helpful in computationally intensive approaches including Density Functional Theory (DFT) and calculations of band structure used for predicting electronic material properties [50,55].

The Total Density of States (TDOS) integrates the influence of all atoms, whereas the Partial Density of States (PDOS) distinguishes the contributions of specific atoms or orbitals, emphasizing their significance in electronic behavior [56]. Variations like these determine how distinct atomic components or orbital contributions impact a material's behavior. In band structure studies, the density of electronic states is essential since it signifies the count of available quantum states across a defined energy interval. At absolute zero, the maximum occupied energy, identified as the Fermi level, possesses a density of states that critically influences electron mobility within a material. Metals with a high DOS at the Fermi level often exhibit enhanced conductivity [50].

Fermi energy is the energy level at absolute zero at which electron occupation is equally likely. Its value is important as it is the cutoff energy level of the electrons, located at the top of the valence band in semiconductors and insulators. The band gap is very important in identifying the ease with which electrons can be excited and thus control electronics and optoelectronics applications. Combined, these values are crucial in deciding whether a material acts as a conductor, a semiconductor, or an insulator [52].

Fermi energy and band gap can be assessed through experimental studies and computational simulations. The analysis of the Fermi energy and band gap involves both experimental techniques and computational approaches. In computation, DFT-based techniques predict band gaps and Fermi energy by evaluating electron density distributions in solids, which aids in classifying materials and predicting electronic properties [54].

1.4.7 Magnetic Behavior and Spin Configurations

Magnetic properties and spin arrangements are central to grasping how materials respond when subjected to external magnetic fields, serving as a cornerstone for their technological applications. Magnetic moments, defined by their alignment and magnitude, are shaped by these conditions, which arise from atomic configuration and electron distribution, as well as interactions between electron spin and electron clouds. A material's magnetism is heavily influenced by the presence of unpaired electrons, and the interactions between them give rise to various magnetic behaviors. These behaviors include ferromagnetism, where aligned moments produce net magnetization, antiferromagnetism, where opposing alignments neutralize magnetization, and paramagnetism, where weak alignment occurs in an external field [49,50].

As a primary property, magnetization reflects the magnetic moment within a unit volume, critical for electromagnets and storage technologies. Another crucial parameter, magnetic susceptibility (χ), measures a material's ease of magnetization in a magnetic field, important for applications needing magnetic responsiveness. Furthermore, coercivity and remanence characterize a material's resistance to demagnetization. The field strength necessary to nullify magnetization is measured by coercivity, whereas remanence shows the magnetization left after the external field is removed. Permanent magnets, which have high coercivity, maintain their magnetization, while soft magnetic materials with low coercivity are easily demagnetized, making them useful in transformer cores [57].

Anisotropy stands for a direction dependence of the magnetic response of a material and forms the very basis of magnetic recording, as it enables finer control over magnetization, and thereby efficient data storage. Temperature influences magnetic properties, weakening magnetic order as it rises, which typically vanishes at specific

critical temperatures: The Curie temperature for ferromagnets and the Néel temperature for antiferromagnets [58].

The atomic structure influences magnetic properties, where metals like iron, cobalt, and nickel display strong magnetism from unpaired electrons, while copper and aluminum, with paired electrons, have only weak magnetism. These properties give insight into the design of magnetic devices for electronic, spintronic, and magnetic storage applications since control of magnetic response is highly related to high performance and reliable operation [59].

The magnetic moment is a vector quantity that represents the strength and direction of a magnetic source. Electric charges, especially electrons, are responsible for a fundamental property of materials. The magnetic moment arises from two main aspects: the intrinsic spin of electrons and their orbital movement around the nucleus. Mathematically, the magnetic moment ($\vec{\mu} = -g \frac{e}{2m} \vec{s}$) can be defined for a single electron. Where g is the Landé g -factor, e electron charge, m electron mass, and $\vec{s}$ is the angular momentum. For a system of multiple magnetic dipoles (like a collection of atoms or molecules), the total magnetic moment is the vector sum of the individual magnetic moments:

$$\vec{M} = \sum_i \vec{\mu}_i \dots\dots\dots (13)$$

Interstitial magnetic moments occur in the empty sites (interstitials) within a crystal structure. These moments can be induced by the surrounding magnetic atoms, impacting the material's overall magnetic properties. Interstitial magnetic moments frequently play a role in phenomena such as magnetoresistance, especially in transition metal alloys. Spin polarization (P) is defined as the difference in the density of states for majority and minority spins at the Fermi level, normalized by the total density of states. It can be mathematically defined as:

$$p = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}} \dots\dots\dots (14)$$

In this context, $n_{\uparrow}$ and $n_{\downarrow}$ represent the densities of states for majority and minority spins, respectively. High spin polarization is useful in spintronics, where it enhances its effectiveness concerning spin transport. Ensuring high polarization benefits the field of

spin transport in gaining high efficiency. Defined by their ability to conduct electricity for one spin channel (majority spins) while remaining insulating for the other (minority spins), half-metals possess a unique property. The electronic band structure is responsible for this phenomenon, which features partially occupied majority spin states and minority spin states that are either vacant or elevated in energy.

Slater-Pauling rules in half-metallic Heusler compounds take total valence electrons as a primary indicator of electronic and magnetic properties. The model allows for predictions about Heusler compound magnetic properties based on the correlated relationship between valence electrons and total spin magnetic moment in a given compound. Both full and quaternary Heusler compounds obey the rule $M_{\text{tot}} = Z_{\text{tot}} - 24$ applies, while half-Heusler compounds follow $M_{\text{tot}} = Z_{\text{tot}} - 18$. These rules, in addition to the orbital hybridization and symmetry rules, are very important for spintronics, as they confirm half-metallicity and help in designing spintronic applications using the beneficial characteristics of Heusler compounds [60].

In closing, magnetic properties derived from atomic and electron configurations are pivotal for fields like data storage and spintronics. By establishing a direct relationship between valence electrons and magnetic moments, the Slater-Pauling rules improve forecasting precision, facilitating the customization of half-metallic Heusler compounds for technological progress.

1.4.8 Optical Transitions and Spectral Properties

Material and compound suitability for technology hinges on their electronic structure. The structure of a material is revealed through its optical characteristics. Absorption, reflectance, and transmittance are among the properties scientists examine to probe this structure. Understanding the electronic band structure is crucial. This includes the band gap and energy states. The band gap is a key parameter. The functionality of semiconductors and insulators in electronic and photonic applications depends largely on the band gap. Devices like LEDs and lasers exhibit improved performance when a direct band gap is present. This is because it enhances light emission [52]. For both photonic devices and renewable energy technologies, optical properties are of vital importance. Effective absorption and emission of light are crucial. These are essential for efficient solar cells, photodetectors, and LEDs. Optimizing absorption is

fundamental for improving solar cell efficiency. Advances in energy storage and conversion are fueled by a deeper understanding of light-material interactions. In photocatalysis, the effectiveness of a material in processes like pollutant degradation and water splitting, using sunlight as an energy source, is determined by its optical absorption properties [61].

In addition to other applications, optical studies provide a non-destructive method for evaluating the quality of materials and identifying any defects or impurities. Analyzing optical spectra allows researchers to detect subtle variations. Optical spectra analysis reveals subtle differences in materials. These differences relate to both structure and composition. This information is essential for quality control. It is particularly important during semiconductor manufacturing [62]. Nanomaterials exhibit unique optical properties due to quantum confinement. This characterization method is particularly effective in studying these materials. The impact of quantum confinement is much greater in nanomaterials compared to bulk material [63]. Consequently, a thorough grasp of optical properties is essential for progress in materials science and technology. It leads to better materials and innovative applications.

DFT calculations offer fundamental insights into the optical properties of materials by describing how electromagnetic radiation interacts with electronic states. Central to this is the complex dielectric function (15) [64].

$$\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega) \dots \dots \dots (15)$$

Describing how the material changes when exposed to electromagnetic radiation. The imaginary component, $\varepsilon_2(\omega)$, directly relates to the absorption processes and is calculated using Fermi's golden rule, expressed as:

$$\varepsilon_2(\omega) = \text{Im } \varepsilon_{ij}(\omega) = \frac{h^2 e^2}{\pi m_e^2 \omega^2} \sum_n \int d^3k \langle \Psi_k^{cn} | \mathbf{p}^i | \Psi_k^{vn} \rangle \langle \Psi_k^{vn} | \mathbf{p}^j | \Psi_k^{cn} \rangle \sigma(E_k^{cn} - E_k^{vn} - \omega) \dots (16)$$

According to Equation (10), transitions occur between valence band states, represented by $|\Psi_k^{vn}\rangle$ with energy E_k^{vn} , and conduction band states, represented by $|\Psi_k^{cn}\rangle$, with energy E_k^{cn} . Here, $\mathbf{p}$ represents the momentum operator, and the matrix elements $\langle \Psi_k^{cn} | \mathbf{p}^i | \Psi_k^{vn} \rangle \langle \Psi_k^{vn} | \mathbf{p}^j | \Psi_k^{cn} \rangle$ represent the transition probabilities between valence (vn) and conduction states (cn).

Kramer's-Kronig relations provide a method for obtaining $\epsilon_1(\omega)$, the real component of the dielectric function, which in turn describes how a material propagates light.

$$\epsilon_1(\omega) = \text{Re}\epsilon_{ij}(\omega) = \sigma_{ij} \frac{2}{\pi} p \int_0^\infty \frac{\omega' \text{Im} \epsilon_{ij}(\omega')}{\omega'^2 - \omega^2} d\omega' \dots\dots\dots (17)$$

This mathematical relationship ensures causality in the material's response and provides a complete description of its linear optical properties. Derivation of various measurable optical parameters, which characterize the material's interaction with light, is possible from these fundamental quantities.

The half-metallic compounds exhibit a dielectric function influenced by two types of transitions. Intra-band transitions result from electron excitation within single bands, while inter-band transitions occur when electrons move between bands. Absorption of electromagnetic radiation is caused by these transitions; the effects from bound electrons are described by $\epsilon(\omega) = \epsilon_{\text{intra}}(\omega) + \epsilon_{\text{inter}}(\omega)$ [65].

Optical behavior analysis hinges on the complex refractive index, $n(\omega)$, made up of the refractive index, $\eta(\omega)$, governing the phase velocity of light in the medium, and the extinction coefficient, $\kappa(\omega)$, describing the material's light absorption capacity.

$$n(\omega) = \eta(\omega) + i\kappa(\omega) \dots\dots\dots (18)$$

The refractive index is given by [66]:

$$\eta(\omega) = \frac{1}{\sqrt{2}} \left((\epsilon_1^2 + \epsilon_2^2)^{\frac{1}{2}} + \epsilon_1 \right)^{\frac{1}{2}} \dots\dots\dots (19)$$

The extinction coefficient exhibits behavior according to:

$$\kappa(\omega) = \frac{1}{\sqrt{2}} \left((\epsilon_1^2 + \epsilon_2^2)^{\frac{1}{2}} - \epsilon_1 \right)^{\frac{1}{2}} \dots\dots\dots (20)$$

Equation (19) establishes a mathematical relationship between the components [67]. These parameters fundamentally determine how electromagnetic waves propagate through and interact with the material.

The mathematical relationship between light attenuation per unit length (quantified by the absorption coefficient) and the dielectric function's real and imaginary components is presented in Equation (21) [68].

$$\alpha(\omega) = \frac{\sqrt{2} \omega}{c} \sqrt{-\epsilon_1(\omega) + \sqrt{\epsilon_1(\omega)^2 + \epsilon_2(\omega)^2}} \dots \dots \dots (21)$$

The absorption coefficient, $\alpha(\omega)$, describes how quickly light intensity decreases within a material. This decrease is measured as light travels through the material. The coefficient is directly related to experimental optical absorption measurements. The absorption coefficient $\alpha(\omega)$ is how fast the light intensity decreases in a material. This is measured as the light goes through the material. The coefficient is directly related to the experimental optical absorption measurements. The absorption coefficient controls light absorption and transmission. These affect the performance of optical coatings. They also affect photodetectors and photovoltaic systems. It measures the amount of light absorbed over a given distance, gives insight into the optical properties of the material. Analyzing a material's absorption coefficient reveals information about its electronic transitions, band structure, and potential for energy and optoelectronic applications. So, it not only predicts optical performance but also guides advanced photonic device design [61].

The complex optical conductivity $\sigma(\omega)$ of a material describes how it conducts electricity in reaction to an oscillating electric field. The real part reflects energy dissipation, whereas the imaginary part accounts for energy storage and phase delay, as expressed in Equation (22):

$$\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega) = -i\epsilon_0\omega(\epsilon(\omega) - 1) \dots \dots \dots (22)$$

The optical reflectivity $R(\omega)$ is the amount of electromagnetic radiation a material reflects at a given frequency. It is determined from the dielectric function $\epsilon(\omega)$, comprising the real part $\text{Re } \epsilon_{ij}$ and the imaginary part $\text{Im } \epsilon_{ij}$, as specified in Equation (23) [66]:

$$R_{ij}(\omega) = \left[\frac{(\text{Re } \epsilon_{ij} + i \text{Im } \epsilon_{ij})^2 - 1}{(\text{Re } \epsilon_{ij} + i \text{Im } \epsilon_{ij})^2 + 1} \right] \dots \dots \dots (23)$$

In solar cell application, the knowledge of reflectivity is central to reducing energy loss through the optimization of light absorption. In thin film technology's production of anti-reflective coatings and mirrors, reflectivity control is also of greatest significance. Reflectivity is thus central to optimizing materials in most photonic and optical technologies.

The energy loss function, $L(\omega)$, is the energy lost by electrons while an electromagnetic wave passes through a material and is computed from the following given equation:

$$L_{ij}(\omega) = -\text{Im} \left\{ \frac{1}{\varepsilon_{ij}(\omega)} \right\} = \frac{\text{Im} \varepsilon_{ij}}{(\text{Re} \varepsilon_{ij})^2 + (\text{Im} \varepsilon_{ij})^2} \dots\dots\dots (24)$$

It is important to note that while DFT provides a robust framework for calculating these optical properties, certain limitations exist. Typically, underestimating bandgaps, standard DFT calculations affect the predicted onset of optical transitions. Many-body effects, such as electron-hole interactions (excitons), are also not captured in the basic formulation. The GW approximation combined with the Bethe-Salpeter Equation (BSE) is often necessary for improved accuracy, especially when excitonic effects significantly influence the optical response in semiconductors and insulators [69].

Essential for the progress of quantum technologies is the study of optical properties. With the rise of quantum computing and communication, understanding and manipulating light-matter interactions at the quantum level becomes increasingly important for developing advanced devices like quantum sensors and secure communication systems.

1.4.9 Thermoelectric Effects and Conductivity

The development of sustainable materials, especially for use in thermoelectric systems, presents a difficult yet essential challenge for scientific progress in industry. Thermoelectric materials have been touted as one of the best prospects to offer energy solutions with an ever-growing range of applications. The effect of thermoelectricity enables waste heat to be converted to electricity, and hence it has applications in energy generation as well as refrigeration. Heavy machines, cars, steel production, and chemical industries generate large quantities of waste heat. In such applications, the effect has special significance. The lost heat can be used to generate electricity or drive refrigeration systems through thermoelectric materials to increase energy efficiency and

reduce waste. Thermoelectrics utilize the principle discovered by Seebeck in 1821. He demonstrated that thermal energy could be converted into electricity. This conversion occurs at the junction of two different metals. Seebeck noted that when a metallic conductor was connected at both ends to two other conductors and subjected to a temperature gradient, a metallic compass needle showed deflection if placed between them. Such observation underlined the connection of heat with electrical potential, setting thereby a premise for further understanding of thermoelectric effects. The electron energy level shifts between the metals created a voltage difference at the junctions, inducing an electric current and thereby producing a magnetic field around the wires. At first unaware of the electric current's presence, Seebeck referred to the phenomenon as the thermomagnetic effect. Further studies indicated that a temperature gradient across different materials leads to an electric potential difference at their junction. Currently, thermocouples use the Seebeck effect primarily for high-precision temperature measurements. The Seebeck coefficient, or thermoelectric power, of a material is defined as [70]:

$$S = - \frac{\Delta V}{\Delta T} \dots \dots \dots (25)$$

where S denotes the Seebeck coefficient, ΔT is the temperature difference along the material, and ΔV is the thermoelectric voltage observed. The minus sign indicates that the voltage gradient is oriented against the temperature gradient.

The Peltier effect refers to a thermoelectric phenomenon where heat is either absorbed or released at the junction of two different conducting materials as an electric current flows through; the current's direction determines the heat exchange process. Jean Charles Athanase Peltier, a French physicist, discovered this effect in 1834 while studying temperature changes at circuit junctions in his experiments. His discovery came just 13 years after Thomas Seebeck had discovered the related Seebeck effect, where a temperature difference between two electrical junctions produces a voltage difference.

At the junction of two dissimilar materials, applying a voltage and allowing current to flow necessitates that charge carriers (electrons or holes) change their energy state to move between the materials. This energy transformation results in heat absorption or release at the junction. To cross the junction in one direction, charge carriers must

absorb heat from their surroundings; conversely, crossing in the opposite direction leads to the release of energy as heat.

According to the equation $Q = \pi I$, the heat transfer rate (Q) at a junction is directly proportional to the current (I), with the Peltier coefficient (π) serving as the proportionality constant. The Peltier coefficient, influenced by the materials used and their temperature, signifies the heat energy transferred per unit charge [71]. In Peltier-effect heat pump devices, several junctions are arranged in series electrically but in parallel thermally. When current is applied, one side cools by absorbing heat, while the other side heats up by releasing heat.

Thermoelectric coolers, also known as Peltier coolers, work by sandwiching arrays of semiconductor pellets between two ceramic plates. When DC passes through the semiconductor array, it creates a temperature difference between the plates; one side gets cold while the other gets hot. Heat is dissipated from the hot side via an attached heat sink and fan; the cold side is utilized for cooling purposes [72].

The Figure of merit (ZT) is a dimensionless parameter expressed as:

$$ZT = \frac{S^2 \sigma T}{\kappa_e + \kappa_l} \dots \dots \dots (26)$$

which quantifies how efficiently a thermoelectric material can convert heat into electricity or vice versa. Electrical conductivity, denoted by σ , quantifies current flow; T represents absolute temperature; and the total thermal conductivity is the sum of electronic (κ_e) and lattice (κ_l) contributions [73].

The power factor, given as ($PF = S^2 \sigma$), measures a material's effectiveness in generating electrical power when exposed to a temperature gradient. A higher power factor suggests efficient electrical behavior, but maximizing overall efficiency also requires careful control of thermal conductivity. The balance between the two is delicate due to their complex relationship.

The maximum efficiency (η_{max}) of a thermoelectric device is described by the equation:

$$\eta_{\max} = \frac{T_H - T_C}{T_H} \frac{\sqrt{1 + ZT} - 1}{\sqrt{1 + ZT} + \frac{T_C}{T_H}} \dots \dots \dots (27)$$

Where T_H and T_C are the hot and cold side temperatures, respectively. This equation shows that efficiency improves with increasing ZT ; however, commercial materials usually have ZT values between 1 and 2 due to practical limitations [74].

The Wiedemann-Franz law, expressed as $\kappa_e = l\sigma T$ (where l is the Lorenz number), establishes a fundamental connection between electrical conductivity and electronic thermal conductivity [75,76]. This relationship presents a significant challenge in optimizing ZT because improving electrical conductivity inevitably increases electronic thermal conductivity, creating a trade-off that limits overall performance.

The primary difficulty in improving ZT by enhancing the power factor or reducing electronic thermal conductivity is the interdependence of these factors. Increasing electrical conductivity enhances the power factor but simultaneously raises electronic thermal conductivity, and efforts to reduce thermal conductivity often reduce electrical efficiency. An inverse relationship exists between the Seebeck coefficient (S) and carrier concentration (n), as well as between electrical conductivity (σ) and effective mass (m^*). Increasing charge carriers improves the electrical conductivity but decreases the Seebeck coefficient [77,78].

The optimization of ZT has basically centered on the reduction of lattice thermal conductivity while at the same time keeping the power factor high. Such actions can be implemented through strategies such as introducing phonon scattering centers, designing nanostructured materials, developing complex crystal structures, and integrating point defects [79].

The electrical conductivity tensor ($\sigma_{\alpha,\beta}$) is mathematically expressed as [80]:

$$\sigma_{\alpha,\beta} = e^2 \sum_{i,k} \left[\frac{-\partial f_o(T, \varepsilon, \mu)}{\partial \varepsilon} \right] v_{\alpha} v_{\beta} \tau_k \dots \dots \dots (28)$$

Where e represents the electron charge, $f_o(T, \varepsilon, \mu)$ denotes the Fermi-Dirac distribution function, v_{α} and v_{β} are the group velocities in the directions α and β , τ_k is the relaxation time, ε is the energy, and μ is the chemical potential. The relationship between a material's microscopic features and its macroscopic electrical conductivity is

captured by this equation, which underscores the significance of carrier velocities ($v_{\square}$, $v_{\square}$) and their relaxation times ($\tau_{\square}$). A comprehensive understanding of these parameters is essential for optimizing thermoelectric performance [81,82].

An effective thermoelectric material should have several critical properties: it needs high electrical conductivity to limit resistive losses, a large Seebeck coefficient to produce substantial voltage from temperature variations, low thermal conductivity to sustain temperature gradients, a suitable band gap for the specific operating temperature, robust mechanical and environmental stability, economical synthesis methods, and a composition that is eco-friendly, utilizing abundant elements.

The creation of high-performance thermoelectric materials requires balancing numerous trade-off properties through new material design and optimization techniques because different requirements interact complexly. This process of optimization is an ongoing process and is driven by evolving methodologies to reveal interdependent properties and discover new material systems with high ZT values coupled with industrial applicability.

Chapter Two

Theoretical Frameworks and Advanced Computational Techniques

Addressing many-body problems through first-principles calculations, this chapter provides a broad perspective on Density Functional Theory (DFT) within a theoretical and computational framework. The study also includes a summary of the necessary theoretical and computational formulations, in addition to software and methods employed for performing the calculations.

2.1 Introduction

Electron interactions dictate material properties at the atomic level. This section explores electron-electron and electron-nuclei interactions. Quantum mechanical calculations predict various properties of quaternary Heusler alloys. Structural, electronic, magnetic, thermal, and optical characteristics define these properties. Electron spin and exchange interactions are governed by the quantum mechanics. This determines the electronic band structure and magnetic characteristics. These are crucial in spintronics. More than any other factor, atomic vibrations (phonons) dictate mechanical characteristics like elasticity and thermal conductivity, emphasizing their role as a quantum effect. For Heusler alloy research, the accurate simulation and prediction of these properties are made possible through quantum mechanical treatment.

A system's complete quantum state is described by its wavefunction, $\Psi(\vec{X}_1, \vec{X}_2 \dots \vec{X}_N, \vec{R}_1, \vec{R}_2 \dots \vec{R}_M)$, a solution to the Schrödinger equation. While analytical solutions exist for simple systems like the hydrogen and helium atoms, the Schrödinger equation becomes computationally intractable for more complex many-body systems. Solving the Schrödinger equation for complex systems is more challenging. The use of appropriate computational tools to solve the quantum mechanical problem allows for the complete determination of material properties. The many-body problem's complexity arises from the number of particles involved and scales exponentially with that number. The existence of the wavefunction in $3N$ dimensions for N particles in 3D presents computational difficulties. Many techniques have been suggested to overcome these. DFT offers an efficient and accurate alternative by considering the three-dimensional electron density. This chapter details the development of DFT, the Kohn-Sham (KS) approach, and the related exchange-correlation approximations.

2.2 The Many-Body Problem

The primary objective of most quantum methods is to find an approximate solution to the time-independent, non-relativistic Schrödinger equation.

$$\hat{H}\Psi(\vec{X}_1, \vec{X}_2 \dots \vec{X}_N, \vec{R}_1, \vec{R}_2 \dots \vec{R}_M) = E\Psi(\vec{X}_1, \vec{X}_2 \dots \vec{X}_N, \vec{R}_1, \vec{R}_2 \dots \vec{R}_M). \dots (29)$$

$\hat{H}$ denotes the Hamiltonian operator for a molecular system of M nuclei and N electrons [83].

$$\hat{H} = \hat{T}_n + \hat{T}_e + \hat{V}_{en} + \hat{V}_{ee} + \hat{V}_{nn} \dots (30)$$

$$\hat{H} = -\frac{\hbar^2}{2} \sum_{i=1}^N \frac{\nabla^2 \vec{R}_i}{M_i} - \frac{\hbar^2}{2} \sum_{i=1}^N \frac{\nabla^2 \vec{r}_i}{m_e} - \frac{1}{4\pi\epsilon_0} \sum_{i,j}^N \frac{e^2 Z_i}{|\vec{R}_i - \vec{r}_j|} + \frac{1}{8\pi\epsilon_0} \sum_{i \neq j}^N \frac{e^2}{|\vec{r}_i - \vec{r}_j|} + \frac{1}{8\pi\epsilon_0} \sum_{i \neq j}^N \frac{e^2 Z_i Z_j}{|\vec{R}_i - \vec{R}_j|} \dots (31)$$

The terms within the Hamiltonian are: the kinetic energy of the nuclei T_n , the kinetic energy of the electrons T_e , the Coulomb interaction between electrons and nuclei V_{en} , the Coulomb interaction between electrons V_{ee} , and the Coulomb interaction between nuclei V_{nn} , and $\hbar$ refers to the reduced Planck constant. For many-body problems, Equation (29), as it describes many-body systems, contains some complicated terms that allow an exact solution only for simple cases like that of a hydrogen atom; therefore, approximations are needed to be solved.

2.3 Born-Oppenheimer Approximation

The Born-Oppenheimer approximation treats nuclei as stationary, given their far greater mass than electrons. Consequently, electronic and nuclear motions are decoupled. The electronic Hamiltonian, simplified in this manner, excludes nuclear kinetic and interaction terms. When the approximation is applied to equation (2), $\hat{T}_n$ is omitted, the final term becomes a constant, and equation (30) is simplified [84].

$$\hat{H} = \hat{T}_e + \hat{V}_{en} + \hat{V}_{ee} + \hat{V}_{nn} \dots (32)$$

The electronic problem is complicated by the fact that interactions between electrons make exact solutions difficult to obtain. As a result, it is necessary to employ additional approximations.

2.4 The variational principle

The variational principle implies that the energy from an approximate wave function $\psi(\vec{r})$ offers an upper bound on the exact ground state energy E_0 . For a system described by the state $|\psi\rangle$, the energy can be estimated by evaluating the expectation value of the Hamiltonian, given by $E[\psi] = \frac{\langle\psi|\hat{H}|\psi\rangle}{\langle\psi|\psi\rangle} \geq E_0$. This principle offers a fundamental and practical approach for approximating the ground state energy of quantum systems.

2.5 Hartree-Fock Theory

Hartree's theory simplifies the complex many-body problem by approximating the motion of electrons in an atom. Each electron moves independently in the averaged field of all other electrons. This is the reduction of the many-body Schrödinger equation to a set of one-electron equations. The state of each electron experiencing the averaged effect of all other electrons is represented by an individual wavefunction; these are combined as a product to form the single-electron wavefunction [85].

$$\Psi_i(\vec{X}_1, \vec{X}_2 \dots \vec{X}_N, \vec{R}_1, \vec{R}_2 \dots \vec{R}_M) = \Psi(\vec{X}_1, \vec{R}_1) \Psi(\vec{X}_2, \vec{R}_2) \dots \Psi(\vec{X}_N, \vec{R}_M) \dots (33)$$

An approximation of the electron-electron term in Equation (32), making it dependent only on the coordinates of a single electron, allows for variable separation while accounting for electron-electron interactions. This Hamiltonian approximation employs an averaged description of interactions. When the Schrödinger equation is solved using this Hamiltonian, it produces optimal single-electron eigenfunctions [86].

$$(T_s + V_{ext} + V_H)\Psi(\vec{r}) = E\Psi(\vec{r}) \dots (34)$$

In this equation, T_s represents the kinetic energy of a single electron, V_{ext} is the external potential, and V_H is the Hartree potential for non-interacting electrons ($\hat{V}_H = \frac{1}{8\pi\epsilon_0} \sum_{i,j}^N \frac{|\Psi(\vec{r}_i)|^2 |\Psi(\vec{r}_j)|^2 d^3r_i d^3r_j}{|\vec{r}_i - \vec{r}_j|}$).

However, this formulation fails to satisfy the Pauli exclusion principle concerning electrons, since it offers a symmetric wavefunction that indicates no two electrons can share identical quantum numbers. The half-integer intrinsic spin of fermions necessitated the development of the Hartree-Fock approximation, which modified the Hartree product wavefunction accordingly.

The foundation of the Hartree-Fock method is the mean field approximation. In this approach, each electron of an N -electron system experiences an average potential field created by the other $N-1$ electrons and the nuclei. A Slater determinant of spin orbitals represents the wavefunction, guaranteeing compliance with the Pauli exclusion principle, is represented as follows:

$$\psi(\vec{x}_1, \vec{x}_2) = \frac{1}{\sqrt{N!}} \begin{bmatrix} \psi_1(\vec{r}_1)|S_1\rangle & \psi_1(\vec{r}_2)|S_2\rangle & \dots & \psi_1(\vec{r}_N)|S_N\rangle \\ \psi_2(\vec{r}_1)|S_1\rangle & \psi_2(\vec{r}_2)|S_2\rangle & \dots & \psi_2(\vec{r}_N)|S_N\rangle \\ \vdots & \vdots & \ddots & \vdots \\ \psi_N(\vec{r}_1)|S_1\rangle & \psi_N(\vec{r}_2)|S_2\rangle & \dots & \psi_N(\vec{r}_N)|S_N\rangle \end{bmatrix} \dots\dots\dots(35)$$

Where $\frac{1}{\sqrt{N!}}$ is a normalization factor that ensures the total wave function is properly normalized for N particles and $|S_N\rangle$ spin wave functions for each electron, where $|S_i\rangle$ is the spin state of electron i . A single Slater determinant represents the approximated many-electron wavefunction in the computationally efficient Hartree-Fock method, which, however, neglects electron correlation. The total Hamiltonian is then separable into N one-electron Hamiltonians. This method is crucial for understanding electronic structure and offers insights into the stability, geometry, and other properties of atoms and molecules [87].

2.6 Density Functional Theory

Density functional theory is a very important calculational tool for ground-state properties of many-body systems. Invented independently by Thomas and by Fermi, this scheme represents an ingenious reduction of computational complexity owing to electron density $\rho(\mathbf{r})$ in such a way that one can carry out calculations in a single-body frame. The total energy $E_{tot}[\rho(\vec{r})]$ as a functional of the electron density $\rho(\mathbf{r})$ can then be expressed in equation (36) [88]:

$$E_{tot}[\rho(\vec{r})] = \frac{3(3\pi^2)^{\frac{2}{3}}}{19} \left[\rho(\vec{r})^{\frac{5}{3}} d^3\vec{r} - z \int \frac{\rho(\vec{r})}{r} d^3\vec{r} + \frac{1}{2} \int \int \frac{\rho(\vec{r}_1)\rho(\vec{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|} d^3\vec{r}_1 d^3\vec{r}_2 \right] \dots\dots\dots(36)$$

The first term corresponds to the kinetic energy of the electron density. In the second term, the interaction energy is illustrated between the electron density and a point charge of magnitude z , which is found at the origin. Finally, the third term quantifies the electron-electron interaction energy, with $\rho(\vec{r}_1)$ and $\rho(\vec{r}_2)$ representing the electron

densities at positions $\vec{r}_1$ and $\vec{r}_2$, respectively. Normalization of the electron density ensures it integrates to the total electron number; N is stated in Equation (37):

$$\int \rho(\vec{r}) d^3\vec{r} = N. \dots\dots\dots(37)$$

Density functional theory rests upon the principle, established by Hohenberg and Kohn, that the electron density fully specifies the ground state properties of a many-body system. At the ground state density, the energy functional is minimized. Electron density, as suggested by Hohenberg and Kohn for the total energy functional, offers a reduced-dimensionality representation of the system, facilitating the calculation of properties like energy and wavefunction [53,55].

$$E_{tot}[\rho(\vec{r})] = F_{HK}[\rho(\vec{r})] + \int \rho(\vec{r})V_{eN}(\vec{r})d\vec{r}. \dots\dots\dots (38)$$

Where V_{eN} is the external potential from the interaction between the electrons and the nuclei and F_{HK} is the Hohenberg and Kohn functional, which is given by equation (39) [89]:

$$F_{HK}[\rho(\vec{r})] = T[\rho(\vec{r})] + V[\rho(\vec{r})] = T_0[\rho(\vec{r})] + T_H[\rho(\vec{r})] + V_{xc}[\rho(\vec{r})] \dots\dots\dots (39)$$

The sum of the exact kinetic energy T [$\rho(\vec{r})$] and the electron-electron interaction energy V [$\rho(\vec{r})$] defines $F_{HK}[\rho(\vec{r})]$. Non-interacting particle kinetic energy is denoted by T_0 , represents the Hartree energy, while V_{xc} , encompassing both exchange and correlation effects, is the exchange-correlation energy. The Hartree-Fock method includes the exchange potential, while the Hartree method does not.

The scheme of non-interacting electrons was made by Kohn and Sham in order to help solve the many-body problem described by Kohn-Sham equations. This equation includes the effective potential containing the external potential $V_{ext}(\vec{r})$, Hartree potential $V_H(\vec{r})$ describing the classic Coulomb interactions and the exchange-correlation potential $V_{xc}(\vec{r})$ as formulated in Equation (40):

$$\left[-\frac{1}{2} \nabla^2 + V_{ext}(\vec{r}) + V_H(\vec{r}) + V_{xc}(\vec{r}) \right] \psi_i(r) = \epsilon_i \psi_i(r) \dots\dots\dots (40)$$

$\psi_i(r)$ represents the orbital wavefunction of the i -th electron, with ϵ_i being its corresponding energy. Crucially, the exchange-correlation potential, $V_{xc}(r)$, accounts

for many-body effects and is the functional derivative of the exchange-correlation energy, $E_{xc}[\rho]$, concerning electron density.

$$V_H = \frac{\delta V_{xc}[\rho(\vec{r})]}{\delta \rho(\vec{r})}. \dots\dots\dots (41)$$

The electron density $\rho(\vec{r})$ is obtained by summing the squares of the Kohn-Sham orbitals:

$$\rho(\vec{r}) = \sum_{i=1}^{occu} \psi_i^*(\vec{r}) \psi_i(\vec{r}) \dots\dots\dots (42)$$

This density, in turn, is used to compute the total energy of the system. The total energy expression in Kohn-Sham DFT is given by:

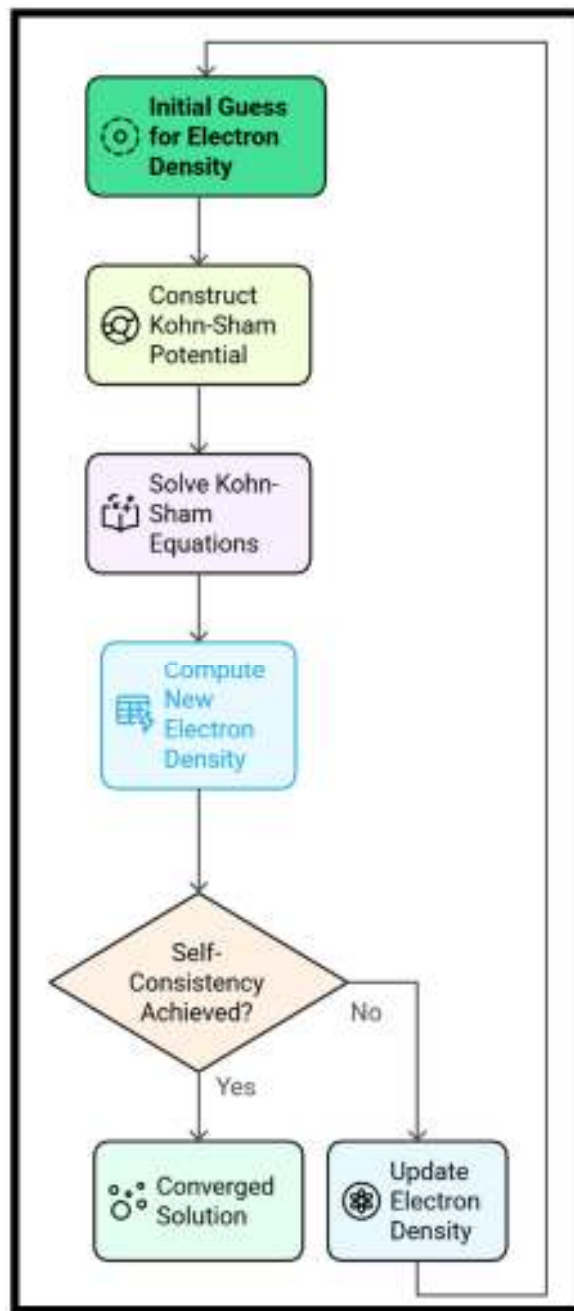
$$E_{tot} = \sum_i \varepsilon_i - \frac{1}{2} \iint \frac{\rho(\vec{r}_1)\rho(\vec{r}_2)}{|\vec{r}_1 - \vec{r}_2|} d^3r_1 d^3r_2 + E_{xc}(\rho) - \int \rho(\vec{r}) \nabla^2 v_{xc}(r) d^3r. \dots\dots (43)$$

The expression for the ground-state energy in DFT comprises the sum of the Kohn-Sham orbital eigenvalues, the classical electron-electron Coulomb interaction energy, the exchange-correlation energy, and a term that corrects for double counting of exchange-correlation effects.

The self-consistency cycle in density functional theory ensures that the input and output electron densities converge, resulting in a stable solution. Through iterative refinements, the process adjusts the electron density until the Kohn-Sham equations yield consistent results across all computational steps. This convergence guarantees accurate calculations of electronic properties, forming the foundation of reliable DFT simulations, as shown in Figure 1.

Figure 1

The iterative framework: achieving convergence in Kohn-Sham DFT



2.7 Exchange Correlation Energy Approximation

Density functional theory encompasses several methods to calculate the exchange-correlation energy by quantitative approximations to the ground-state energy. The most important ones are the local density approximation (LDA) and generalized gradient approximation (GGA), which both see widespread use in applications.

2.7.1 Local Density Approximation

In the Local Density Approximation (LDA), the exchange-correlation energy, represented as E_{xc}^{LDA} , is determined exclusively by the local electron density $\rho(r)$, based on the premise of a homogeneous electron gas at that particular location. The dependency is specified as follows [90,91]:

$$E_{xc}^{LDA} = \int \rho(r) \varepsilon_{xc}(\rho(r)) dr. \dots\dots\dots (44)$$

$$\varepsilon_{xc}(n(\vec{r})) = \varepsilon_x(n(\vec{r})) + \varepsilon_c(n(\vec{r})). \dots\dots\dots (45)$$

The exchange energy is represented by $\varepsilon_x(\rho(\vec{r}))$, a solvable term describing the exchange interaction of an electron within a homogeneous electron gas. LDA is a good starting point for simple systems with uniform electron densities, but its limitations in handling more complex materials necessitate the use of generalized gradient approximation (GGA), hybrid functionals, or other corrections for more accurate predictions.

2.7.2 Generalized Gradient Approximation

The Generalized Gradient Approximation GGA enhances the Local Density Approximation LDA in Density Functional Theory DFT by incorporating the gradient of the electron density, thereby addressing the limitations of LDA in systems with varying densities [92,93].

$$E_{xc}^{GGA} = \int \rho(r) \varepsilon_{xc}(\rho(r)) F(|\nabla \rho(r)|) dr \dots\dots\dots (46)$$

LDA energy density is modified in the GGA by incorporating the gradient via a gain function. This approach succeeds where the initial GEA failed. A common GGA functional, PBE, is known for its accuracy and ease of use. However, it shares with LDA a difficulty in accurately representing band gaps of semiconductors and the complexities of highly correlated systems. Material and semiconductor band gap calculations challenge LDA and PBE. Exchange-correlation energy is very important for DFT to properly represent electron interaction. The Modified Becke-Johnson (mBJ) method is an improvement over GGA functionals for material science.

2.7.3 The Modified Becke-Johnson Potential

The Modified Becke-Johnson (mBJ) potential improves prediction through electron density and its gradient. Parameterized forms strive for more accuracy. Exchange potential is improved through use of electron density and kinetic energy density in solving LDA and GGA shortcomings. The parameters involved are derived empirically to ensure that the calculated band gaps are closer to experimental values. A correction to the exchange-correlation potential, the TB-mBJ potential is applied in DFT calculations after an initial LDA or GGA calculation. By iteratively applying this correction, the band structure undergoes refinement, enhancing band gap accuracy in semiconductors and insulators [94]. Consequently, WIEN2K and similar codes incorporate this approach with ease [95].

2.7.4 Augmented Plane Wave (APW) method

In the APW method, electron behavior is defined by differentiating between two distinct regions: the first being close to atomic nuclei, where strong Coulomb potentials compel electrons to behave like bound atomic electrons, and the second being in interstitial regions, where they exhibit attributes of free electrons. Muffin-tin spheres define specific regions as non-overlapping spherical zones centered on atomic nuclei. Inside these spheres, the wave function is expressed as a combination of radial solutions to the Schrödinger equation and spherical harmonics, whereas in the interstitial region, a plane-wave basis is applied. The plane waves that form the basis of the APW method, represented $\phi_{\vec{K}}^{\vec{k}}(\vec{r}, E)$, are described by equation (47) [96,97].

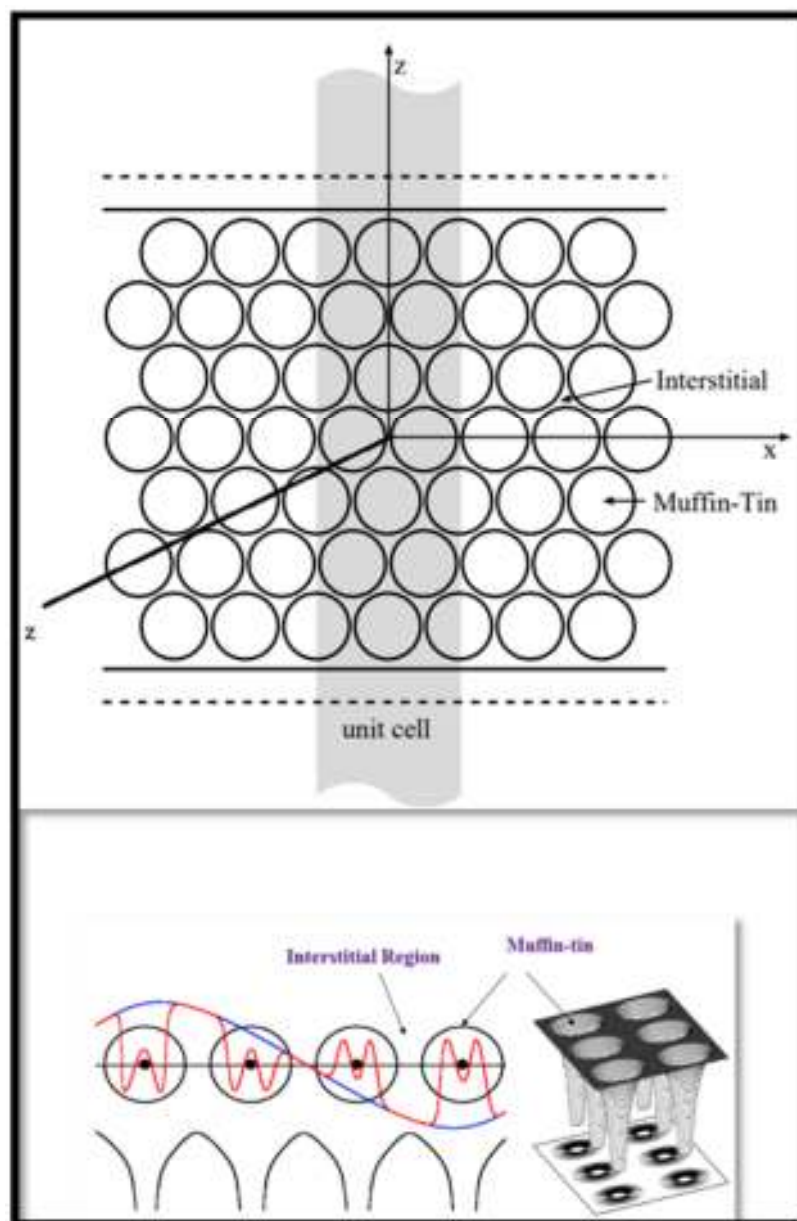
$$\phi_{\vec{K}}^{\vec{k}}(\vec{r}, E) = \begin{cases} \frac{1}{\sqrt{V}} e^{i(\vec{k}+\vec{K}) \cdot \vec{r}} & \vec{r} \in I \\ \sum_{l,m} A_{lm}^{\alpha, \vec{k}+\vec{K}} u_l^{\alpha}(r', E) Y_m^l(\hat{r}') & r' < S_{\alpha} \end{cases} \dots\dots\dots (47)$$

where $\vec{k}$ is the electron wave vector, $\vec{K}$ a reciprocal lattice vector, $\vec{r} \in I$ interstitial region, and V is the crystal volume. Inside the atomic spheres $r' < S_{\alpha}$, the wave function is expanded in terms of spherical harmonics $Y_m^l(\hat{r}')$ and radial functions $u_l^{\alpha}(r', E)$, with coefficients $A_{lm}^{\alpha, \vec{k}+\vec{K}}$ that depend on the angular momentum quantum numbers and atomic properties. For each region in this method, the construction of wave functions varies. Within the muffin-tin spheres, they take the form of linear combinations of spherical harmonics and radial functions that are based on the solutions

to the radial Schrödinger equation. Unlike in the muffin-tin spheres, wave functions in the interstitial space are constructed as linear combinations of plane waves, as depicted in Figure 2. Their smooth transition at the sphere boundaries is a critical factor in establishing the coefficients in the mathematical description. The APW method's computational burden arises from the need to evaluate radial functions at unknown band energies, leading to a nonlinear eigenvalue problem that requires iterative diagonalization.

Figure 2

Schematic representation of the APW



2.7.5 The Linearized Augmented Plane Wave Method

The Linearized Augmented Plane Wave (LAPW) method is a highly accurate approach in density functional theory DFT for solving the Kohn-Sham equations, particularly effective for crystalline solids and complex materials. Electronic wave functions are represented accurately by this full-potential method, capturing both their rapid variation near atomic nuclei and smoother interstitial behavior within the crystal lattice. This allows the method to treat core and valence electrons with high precision. In Andersen's LAPW approach [96] the energy dependence of the radial wave functions $u_l(r, E_l)$ within atomic spheres is expanded by utilizing their energy derivatives.

$$\frac{\partial u^\alpha(r', E)}{\partial E} = \dot{u}^\alpha(r', E). \quad (48)$$

A linear combination of radial functions multiplied by spherical harmonics forms the basis of this approach. The regular solution of the radial Schrödinger equation at a specific energy level determines the use of spherical harmonics $Y_{lm}(r)$. Inside the sphere, the spherical component of the potential, $u_l(r, E_l)$, is represented by the energy derivative of u_l at the same energy level E_l , denoted by $\dot{u}^\alpha(r', E)$ [98].

$$\phi_{\vec{K}}(\vec{r}, E) = \sum_{l,m} (a_{lm}^{\alpha, \vec{K}+\vec{K}} u_l^\alpha(r', E) + b_{lm}^{\alpha, \vec{K}+\vec{K}} \dot{u}_l^\alpha(r', E)) Y_m^l(\hat{r}') \quad r' < S_\alpha \quad (49)$$

Within the interstitial space, the approach of plane wave expansion is adopted.

$$\phi_{\vec{K}}(\vec{r}, E) = \frac{1}{\sqrt{V}} e^{i(\vec{K}+\vec{K}) \cdot \vec{r}} \quad \vec{r} \in I. \quad (50)$$

In its standard representation, the LAPW method defines the potential in this way:

$$V(\vec{r}) = \begin{cases} \sum_{lm} V_{lm}(r) Y_{lm}(\hat{r}) & \text{for } r' < S_\alpha \\ \sum_{\vec{K}} V_{\vec{K}} e^{i\vec{K} \cdot \vec{r}} & \text{for } \vec{r} \in I \end{cases} \quad (51)$$

The LAPW method requires matching these functions and their derivatives at the boundary of the MT spheres, ensuring continuity. Notably robust for all-electron calculations, this method does not require pseudopotentials, thus ensuring precision for both valence and core electron states in complex systems (all electron system).

2.8 The WIEN2k code

The WIEN2k code, first developed in 1990 by P. Blaha, K. Schwartz, P. Sorintin, and S.B. Trickey from the Institute of Materials Chemistry at Vienna University of Technology, has been subject to frequent updates and improvements [96]. These revisions are reflected in versions labeled by release year, including WIEN93, WIEN95, WIEN97, 2014 WIEN2K and others. For this work, we utilized the 2024 WIEN2k version to examine three quaternary Heusler structures. This simulation code is mainly written in FORTRAN90, runs on the LINUX operating system, and integrates a series of standalone programs connected through C-SHELL scripts. Using the APW+lo method, this code provides a highly versatile computational approach to solving the Kohn-Sham equations within DFT. WIEN2k, using a self-consistent and complete potential treatment, provides enhanced accuracy for modeling complex systems like quaternary Heusler alloys.

The core advantage of WIEN2k lies in its adaptability to a wide range of electronic structure tasks, including band structure calculations and specialized features such as NMR shielding tensors and electric polarization. APW+lo method in WIEN2k combines augmented plane waves in interstitial spaces and radial wave functions within atomic spheres. Linearized APW is extended by including local orbitals for faster convergence and accuracy in complex electronic structures. The method is good at handling semi-core electrons and localized electrons that problematize simple approaches. WIEN2k's modular nature structures individual programs using shell scripts, allowing geometry optimization, band structure calculation, and charge density representation through flexible implementations. The program accommodates multiple exchange-correlation functionals, with flexible adaptation to various materials. Users handle calculations and visualize through a web-based interface (w2web), while retaining parameters' control (wien2k). Indeed, WIEN2k demands experienced convergence management using its Self-Consistent Field (SCF) cycle with iteratively repeated runs in order to achieve stable electron and potential fields. SCF stabilizes numerics using diverse convergence criteria and facilitates parallelization to make it more efficient for large-scale implementations. This arrangement ensures high accuracy and reproducibility, necessary to investigate complex materials such as quaternary Heusler alloys (WIEN2k) [96].

2.9 BoltzTraP Code: A Computational Tool for Transport Properties

BoltzTraP calculates band structure-dependent properties, especially semi-classical transport coefficients, using Boltzmann transport theory [99]. A Fourier-smoothed interpolation approach is utilized to derive analytical electronic band descriptions, which support the extraction of essential transport property derivatives. Success has been validated for this framework across multiple material systems from thermoelectric modules to intermetallic phases and high-critical-temperature superconductors [99].

The core of BoltzTraP's methodology is a Fourier expansion of band energies, which maintains space group symmetry through star functions. This approach guarantees physically consistent band structure models by carefully minimizing energy fluctuations between discrete data points and adhering closely to calculated band energies. These facilitate efficient derivation of thermoelectric properties such as conductivity tensors, Seebeck responses, and electronic heat transport metrics.

BoltzTraP is highly compatible with density functional theory (DFT) codes like WIEN2k, requiring only crystal structure and eigen-energies as input, making it easy to interface with other band-structure codes [99]. Low computational costs enable BoltzTraP's application to extensive material datasets, whereas its interpolation of refined k-meshes and derivation of higher-order gradients ensure rigorous transport evaluations. For thermoelectric and electronic structure research, these dual strengths solidify its position as a leading tool.

2.10 The Slack Model for Lattice Thermal Conductivity

The Slack model critically estimates lattice thermal conductivity (κ_{lat}) in crystalline materials, particularly semiconductors and insulators, where phonon-dominated heat transport prevails. This section systematically explores the model's mathematical foundation and its role in predicting heat transport properties. According to the model, the efficiency of heat conduction is largely dictated by phonon scattering phenomena, particularly those resulting from anharmonic lattice vibrations and phonon-phonon interactions.

To determine the lattice thermal conductivity (κ_l) of the alloy, the Slack model was applied, and its mathematical representation is given in the equation that follows [100,101]:

$$\kappa_l = A \frac{M_a \theta^3 \delta}{\gamma^2 T n^{2/3}} \dots \dots \dots (52)$$

The following calculation determines A, the Slack constant, which serves as a coefficient in Equation (52).

$$A = \frac{2.43 \times 10^{-8}}{1 - \frac{0.541}{\gamma} - \frac{0.228}{\gamma^2}} \dots \dots \dots (53)$$

The thermal conductivity of materials exhibits a substantial dependence on the mean atomic mass (M_a). The volume each atom occupies in a unit cell is quantified as atomic volume (δ), and lattice vibration behavior is described by the Debye temperature (θ_D). In a unit cell, n corresponds to the number of atoms, and T indicates the absolute temperature. The Grüneisen parameter (γ), which lacks dimensions, assesses the influence of volume variations on lattice vibrational behavior [101].

$$\gamma = \frac{9 - 12 \left(\frac{V_t}{V_l} \right)^2}{2 - 4 \left(\frac{V_t}{V_l} \right)^2} \dots \dots \dots (54)$$

The transverse elastic wave velocity (V_t) and longitudinal elastic wave velocity (V_l) are key parameters in the analysis. The Grüneisen parameter (γ) and Debye temperature (θ_D) were determined through calculations performed with the WIEN2k software, utilizing its IRelast module [36,102]. Widely recognized for its effectiveness, the Slack model enables the estimation of lattice thermal conductivity in crystalline materials. Even though it does not comprehensively consider defect structures or electronic contributions, the Slack model significantly aids research in thermal regulation and energy applications. Its predictive capability, based on fundamental material characteristics, makes it particularly beneficial for initial assessments of thermoelectric and heat-management materials.

2.11 Computational Methodology

The quaternary Heusler alloys *FeMnCrGe*, *FeMnScAl*, and *FeMnScGa* were precisely examined for their structural, electronic, elastic, optical, and thermoelectric properties using a computational methodology grounded in Density Functional Theory (DFT) and implemented in WIEN2k [96]. Structural optimizations driven by the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional integrated within the GGA architecture were systematically conducted, complemented by FP-LAPW simulations celebrated for their precision in capturing material complexities [20,22,23].

By employing Murnaghan's equation of state [28], unit cell volumes were relaxed to accurately compute lattice constants and bulk moduli. Iterative steps adhered to rigorous convergence benchmarks, stopping only when total energy differences dropped beneath 10^{-5} Ry and atomic forces were reduced below 10^{-5} Ry/Bohr, ensuring reliable numerical results.

Recognizing the inherent limitations of standard GGA in predicting bandgaps, the modified Becke-Johnson (mBJ-GGA) potential was employed to enhance the accuracy of electronic band gap and density of states (DOS) calculations [94,105,106]. Each compound crystallized in the cubic structure (space group F-43m, #216), with muffin-tin radii optimized to avoid overlap while maximizing enclosed volume. For *FeMnCrGe*, atomic sphere radii of 2.20 *a.u.* were assigned to *Fe* and *Mn*, and 2.15 *a.u.* to *Cr* and *Ge*. In *FeMnScAl*, radii were set at 2.38 *a.u.* for *Fe* and *Mn*, 2.32 *a.u.* for *Sc*, and 2.20 *a.u.* for *Al*. For *FeMnScGa*, the values were 2.43 *a.u.* for *Fe* and *Mn*, and 2.31 *a.u.* for *Sc* and *Ga*. A 12×12×12 Monkhorst-Pack k-point grid was used for Brillouin zone sampling, ensuring robust convergence with 2000–3000 k-points in the full zone and 165–375 k-points in the irreducible zone. In Table B.9 convergence testing was carried out by systematically increasing the number of k-points, while the R_{KMAX} parameter was maintained at a fixed value of 8, to ensure that the computed total energy and other derived physical properties of the crystalline materials were accurately and reliably determined.

To calculate the elastic properties C_{11} , C_{12} , and C_{44} , strains ranging from –3% to +3% were applied to optimized structures at zero pressure and 0 K. Polynomial fitting of the energy-strain data allowed the extraction of elastic constants as second

derivatives of energy concerning strain. Using the cubic-elastic package, mechanical stability and anisotropic behavior were confirmed [36,102]. Spin-polarized SCF calculations identified metallic characteristics in the spin-up channel and semiconducting traits in the spin-down channel, affirming the compounds' half-metallic nature. For FeMnScGa, partial charges and orbital-resolved DOS were analyzed using the `x lapw2 -qtl` command, while `x-tetra` facilitated Brillouin zone integration. The DOS plot utility provided detailed visualizations of atomic contributions to electronic states.

The joint density of states (JDOS) approach, a fundamental concept in solid-state physics, describes the number of available electronic transitions between two energy bands. Using this approach, the optical properties were investigated through the WIEN2k optics module [107,108]. Plasma frequencies were obtained from the `compound.outputjoint` file after configuring the `injoint` parameter. By applying a 10,000 k-point mesh and including Drude contributions for free-carrier effects, high accuracy was achieved. The dielectric function and absorption coefficient were thoroughly determined via Kramers-Kronig transformations, yielding significant optical data [109]. Thermoelectric coefficients, including the Seebeck coefficient (S) and electrical conductivity (σ/τ), were determined with BoltzTraP under the rigid-band approximation [99]. Eigenvalues for both spin channels utilized a 10,000 k-point mesh, and SCF results supplied the Fermi energies and carrier densities. These analyses offered valuable insights into the thermoelectric and spintronic applications of the compounds.

Chapter Three

Result and discussion

This chapter provides a complete investigation into the fundamental properties of three quaternary Heusler compounds: *FeMnCrGe*, *FeMnScAl*, and *FeMnScGa*. The study, conducted via systematic density functional theory-based first-principles calculations, evaluates their applicability in spintronics and green energy sectors. Included in the study are structural optimization, elastic stability analysis, electronic band structure calculations, magnetic ordering characteristics, optical response evaluation, thermoelectric performance assessment, and phonon dynamics investigation. The influence of elemental substitution on these properties is examined, and structure-property relationships vital for materials design are established. Distinctive features that make each compound suitable for specific technological applications are revealed in our findings, which are discussed in detail throughout this chapter.

3.1 FeMnCrGe Compound

3.1.1 Formation Energy and Structural Properties

The thermodynamic stability of the *FeMnCrGe* compound was determined by work on the formation and cohesive energies [110,111]. The formation energy, $E_{Form.}$, is one important measure for the ground-state stability of a compound. It describes the energy required to break the compound into neutral free atoms [112]. In this respect, the negative value of the formation energy for *FeMnCrGe*, as obtained in Table 1, points toward its thermodynamic stability [112,113]. The formation energy, $E_{Form.}$, of the compound has been estimated using Equation (52) [30,110]:

$$E_{Form.} = E_{FeMnCrGe(x)}^{Cubic} - (E_{Fe}^{Cubic} + E_{Mn}^{Cubic} + E_{Cr}^{Cubic} + E_{Ge}^{Cubic}) \dots\dots\dots (55)$$

In this context, $E_{FeMnCrGe(x)}^{Cubic}$ signifies the minimum energy for type x (where $x = 1,2,3$) of the compound. The minimum energies of the constituent elements *Fe*, *Mn*, *Cr*, and *Ge* are given by E_{Fe}^{Cubic} , E_{Mn}^{Cubic} , E_{Cr}^{Cubic} and E_{Ge}^{Cubic} .

In the present study, the cohesive energy (E_{coh}) for all three kinds of *FeMnCrGe* compounds is calculated by subtracting the total energy of the compound from the sum

of the energies of its constituent atoms in their isolated states, and then dividing by the total number of atoms in the unit cell [114]:.

$$E_{coh.} = \frac{(E_{Fe}^{iso} + E_{Mn}^{iso} + E_{Cr}^{iso} + E_{Ge}^{iso}) - E_{FeMnCrGe(x)}^{Cubic}}{4} \dots\dots\dots (56)$$

In this case, E_{Fe}^{iso} , E_{Mn}^{iso} , E_{Cr}^{iso} , and E_{Ge}^{iso} refer to the isolated atomic energies of Fe, Mn, Cr, and Ge atoms, respectively. Meanwhile, $E_{FeMnCrGe(x)}^{Cubic}$ represents the total energy per formula unit of the corresponding *FeMnCrGe* compound.

The stability of crystal structures is assessed by cohesive energy calculations, where positive values denote stable configurations. In Table 3, the cohesive energy values for all three variants of the *FeMnCrGe* compound are presented. Together with mechanical stability analysis, these findings confirm the compounds' potential for both future research and practical applications.

The *FeMnCrGe* compound is characterized by a *LiMgPdSn*-type face-centered cubic crystal structure and is categorized under space group Fm-43 (number 216). Its structural stability evaluation involves three atomic configurations shaped by the placement of its elements. *Ge* atoms are fixed at the *4d* Wyckoff position (0.75, 0.75, 0.75), and *Fe*, *Mn*, and *Cr* atoms are positioned at *4a* (0, 0, 0), *4c* (0.25, 0.25, 0.25), and *4b* (0.5, 0.5, 0.5) sites, as detailed in Table 1 [115–117]. Structural characterization employed the Murnaghan Equation of State, expressed in Equation (57), which relates total energy to volume per formula unit through key parameters: equilibrium volume (V_0), bulk modulus (B), and its derivative (B'), and minimum energy at 0 K (E_0) [28,118].

$$E(V) = E_0(V) + \frac{BV}{B'} \left[\frac{\left(\frac{V_0}{V}\right)^{B'}}{B'-1} + 1 \right] - \frac{BV_0}{B'-1} \dots\dots\dots (57)$$

The bulk modulus (B), pressure (P), and volume (V) are interconnected through the following equation: $B = -V \frac{dP}{dV} = V \frac{d^2E}{dV^2}$, where $P = -\frac{dE}{dV}$.

Structural configurations were analyzed by a volume optimization method based on the Murnaghan equation of state, and their results are shown in Figure 3. Among the three examined atomic configurations, type 1 has the lowest energy at zero pressure, making

it the most stable form of *FeMnCrGe*. Therefore, this configuration is recognized as the most energetically favorable for the compound under study. Table 1 tabulates the computed equilibrium structural parameters: optimized energy (E_0), lattice parameter (a_0), bulk modulus (B), and its derivative (B') obtained from volume optimization and fitted to the equation of state. From this point forward, all findings and discussions in this paper will focus exclusively on the type 1 structure, as it has been identified as the most stable configuration. Three distinct structural types were considered in constructing the compound's crystalline structure, which are illustrated in Figure 4.

The variations in structural rigidity for the atomic configurations of the *FeMnCrGe* alloy are presented in Table 1. With its elevated bulk modulus, the type 1 configuration exhibits improved compression resistance, affirming its greater structural stability. In contrast, the type 2 configuration exhibits a moderate resistance to compression, whereas the type 3 structure, with its lower bulk modulus, shows increased compressibility under pressure, indicating reduced mechanical stability. These variations emphasize the critical role of atomic arrangement in determining the alloy's mechanical properties [37].

Figure 3

Volume optimization results for FeMnCrGe configurations

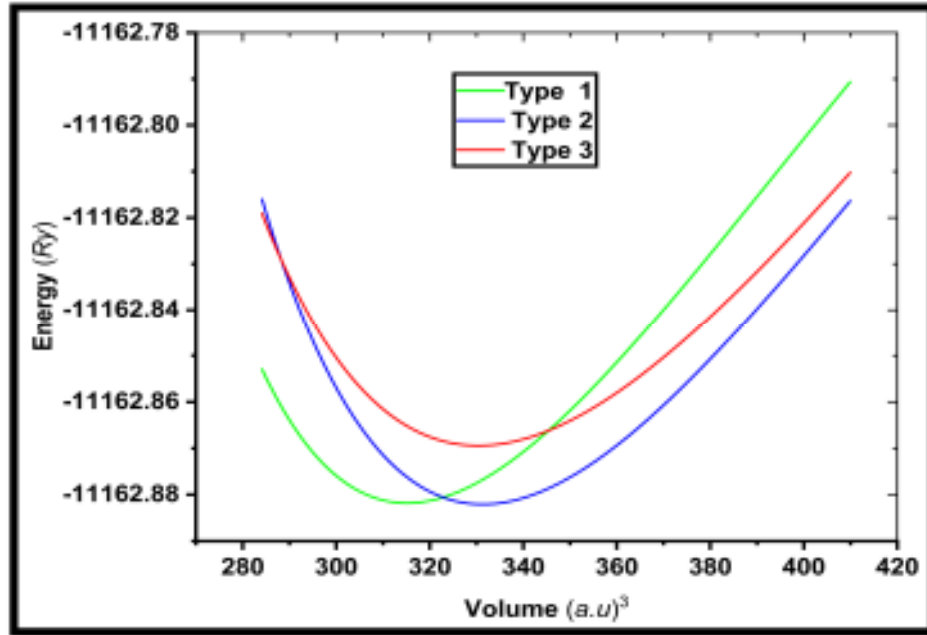


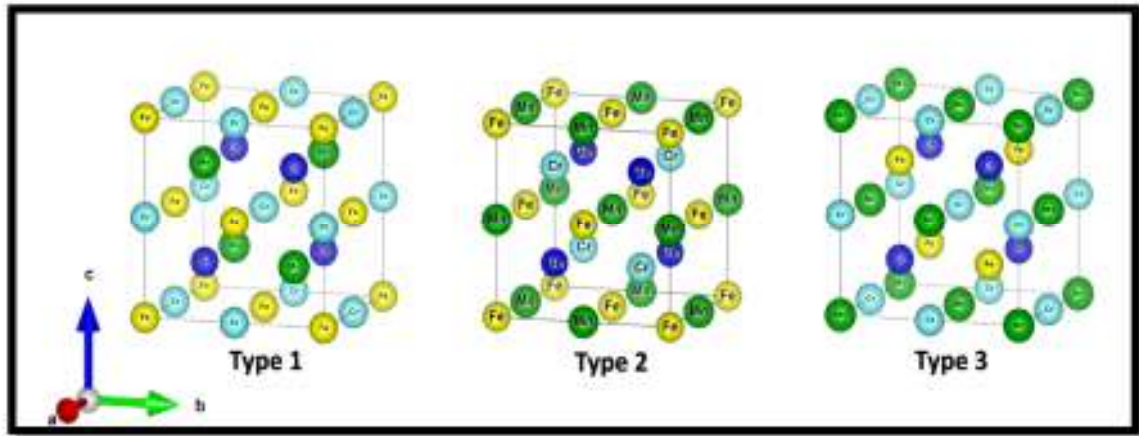
Table 1

The optimized lattice parameter a ($\text{\AA}$), bulk modulus B (GPa), and its pressure derivative B' (GPa) for FeMnCrGe

Property	Type 1	Type 2	Type 3
Fe Position	(0, 0, 0)	(0, 0, 0)	(1/4, 1/4, 1/4)
Mn Position	(1/4, 1/4, 1/4)	(1/2, 1/2, 1/2)	(0, 0, 0)
Cr Position	(1/2, 1/2, 1/2)	(1/4, 1/4, 1/4)	(1/2, 1/2, 1/2)
Ge Position	(3/4, 3/4, 3/4)	(3/4, 3/4, 3/4)	(3/4, 3/4, 3/4)
Lattice Parameter a ($\text{\AA}$)	5.7113	5.7137	5.8058
Ground state energy E_0 (Ry)	-11162.882	-11162.881	-11162.869
B'	10.6409	7.6153	6.9354
Bulk Modulus B (GPa)	220.858	192.121	153.801
Formation Energy (Ry)	-5.9572	-5.9568	-5.9443
Cohesive Energy (Ry/Atom)	2.5003	2.4996	2.511

Figure 4

Three Crystal Structures of FeMnCrGe Compound



3.1.2 Elastic Properties

A material's mechanical stability, essential for practical applications, is fundamentally determined by its elastic properties. This study thoroughly evaluates the mechanical properties of QHA FeMnCrGe at zero pressure, analyzing parameters such as elastic constants (C_{ij}), bulk modulus (B), shear modulus (S), bulk-to-shear modulus ratio (B/S), Poisson's ratio (ν), Young's modulus (Y), and anisotropy factor (A). The criteria for mechanical stability in conventional elasticity require that the conditions $C_{11} > 0$, $C_{11} +$

$2C_{12} > 0$, $C_{11} - C_{12} > 0$, and $C_{44} > 0$ be satisfied [119]. Table 2 presents a comprehensive compilation of mechanical parameters for QHA FeMnCrGe, calculated under specific conditions: 0 K temperature, zero pressure, and zero strain (δ) [36,102]. The results of the computations confirm that the FeMnCrGe type 1 configuration is mechanically stable. The mechanical and bonding properties of the QHA FeMnCrGe type 1 crystalline structure, specifically its bulk and shear moduli in response to external forces, are investigated in this analysis [37]. Young's modulus determines a material's stiffness, classifying it as rigid (higher values) or flexible (lower values) [120]. The material's stress response, stability, and bonding characteristics can be understood through its Poisson's ratio. A Poisson's ratio of 0.26 serves as a critical boundary; values above this indicate ductility, while those below signifies brittleness [38]. The Poisson's ratio, at 0.229 and below the 0.26 threshold, confirms the brittle nature of the material. The Poisson's ratio, calculated as 0.229 and below the 0.26 threshold, indicates the material's brittleness. The material's brittleness is further confirmed by its B/S ratio of 1.51772, falling below the 1.75 ductility criterion [120]. Predominantly covalent bonding is indicated when the Poisson's ratio is below 0.25, whereas predominantly ionic bonding is indicated when the ratio falls between 0.25 and 0.50. A Poisson's ratio of 0.229 confirms the material predominantly exhibits covalent bonding [44]. The compound exhibits anisotropic behavior, with $A \neq 1$. Consequently, its mechanical properties are direction-dependent [37,47]. Measured elastic wave velocities: transverse, 4309.45 m/s; longitudinal, 7145.56 m/s; average, 4764.89 m/s. The calculated Debye temperature (Θ_D) is 627.139 K, and the estimated melting point falls between 2351.3821 K and 2951.3821 K [48,121]. According to these findings, FeMnCrGe type 2 shows a combination of mechanical stability and brittleness, making it ideal for use in environments that demand exceptional resistance to both mechanical stress and heat.

Table 2
Elastic Properties of FeCrMnGe Quaternary Heusler

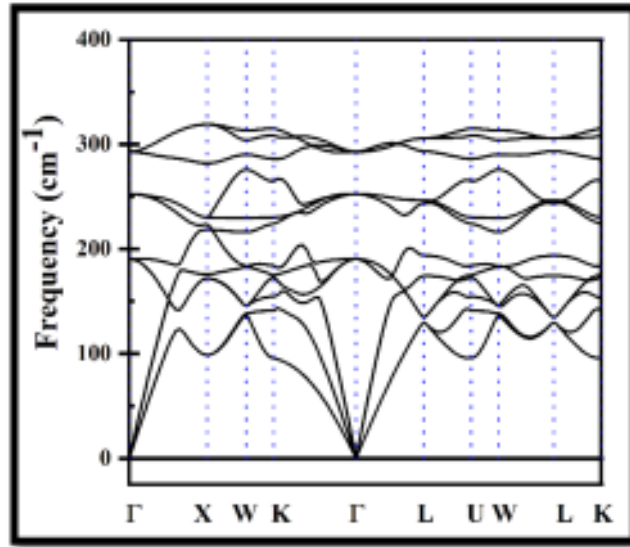
Mechanical Parameters	Unit	<i>FeCrMnGe</i> Type 1
C_{11}	<i>GPa</i>	355.05
C_{12}	<i>GPa</i>	155.54
C_{44}	<i>GPa</i>	212.36
Shear Modulus (S_R)	<i>GPa</i>	146.3
Bulk Modulus (B)	<i>GPa</i>)	222.047
Young's Modulus (Y)	<i>GPa</i>	359.871
B/S_R	-	1.5177
Poisson's Ratio (ν)	-	0.229
Anisotropic Factor (A)	-	2.1288

3.1.3 Phonon Dispersion Analysis

Figure 5 provides a detailed picture of FeMnCrGe type 1's phonon dispersion relation, thus detailing the vital information of vibrational modes to analyze its structure's stability. The present study supports the material's vibrational stability. In addition to that, the lack of imaginary modes on the Brillouin zone's path (Γ -X-W-K- Γ -L-U-W-L-K) supports the proof of such a structure's dynamic stability. The distinct phonon modes are represented by each dispersion curve, which is linked to atomic vibrations within the crystal lattice. These curves reveal how vibrational frequencies shift with changes in the wavevector, providing essential insights into atomic interactions and the overall stiffness of the lattice. The absence of negative frequencies suggests that atomic displacements are counteracted by restoring forces, helping maintain equilibrium. The counteraction of atomic displacements by restoring forces, as indicated by the lack of negative frequencies, ensures equilibrium maintenance [122].

Figure 5

FeMnCrGe's phonon dispersion curve along high-symmetry paths in the Brillouin Zone



3.1.4 Electronic Properties

A complete understanding of chemical bonds in compounds and alloys is achieved through the investigation of a material's electronic properties. The analysis reveals the basic interactions among the elements. Following this, the electronic properties of FeMnCrGe type 1 are explored to gain a deeper understanding of its electronic behavior. The electronic structure of FeMnCrGe is explained based on calculated total and partial densities of states (DOS), using both Generalized Gradient Approximation (GGA) and mBJ exchange potential. The calculations are vital in understanding factors that lead to energy band gaps, as seen in Figures 6(a-e) and 7(a-e). The figures show the total and partial densities of states of the quaternary Heusler compound FeMnCrGe. It has been found that the largest contribution to DOS near the Fermi level is due to Fe's d-orbitals, Cr's d-orbitals, and Mn's d-orbitals that hugely contribute to both the valence and conduction bands. There is also a small contribution due to p-orbitals and s-orbitals of the elements in both channels of spin. Furthermore, the s-orbitals and p-orbitals of Ge also provide small contributions in both the valence and conduction bands, as observed in both approximation methods. Analysis reveals that QHA FeMnCrGe is close to half-metallic. It also achieves full spin polarization. The absence of spin-down states near the Fermi level results in the electronic structure being shaped by spin-up states. The substantial spin polarization at the Fermi level positions FeMnCrGe as an ideal material for spintronic uses.

Figure 6

Total and Partial Density of States (DOS) for FeMnCrGe Using GGA Approximation

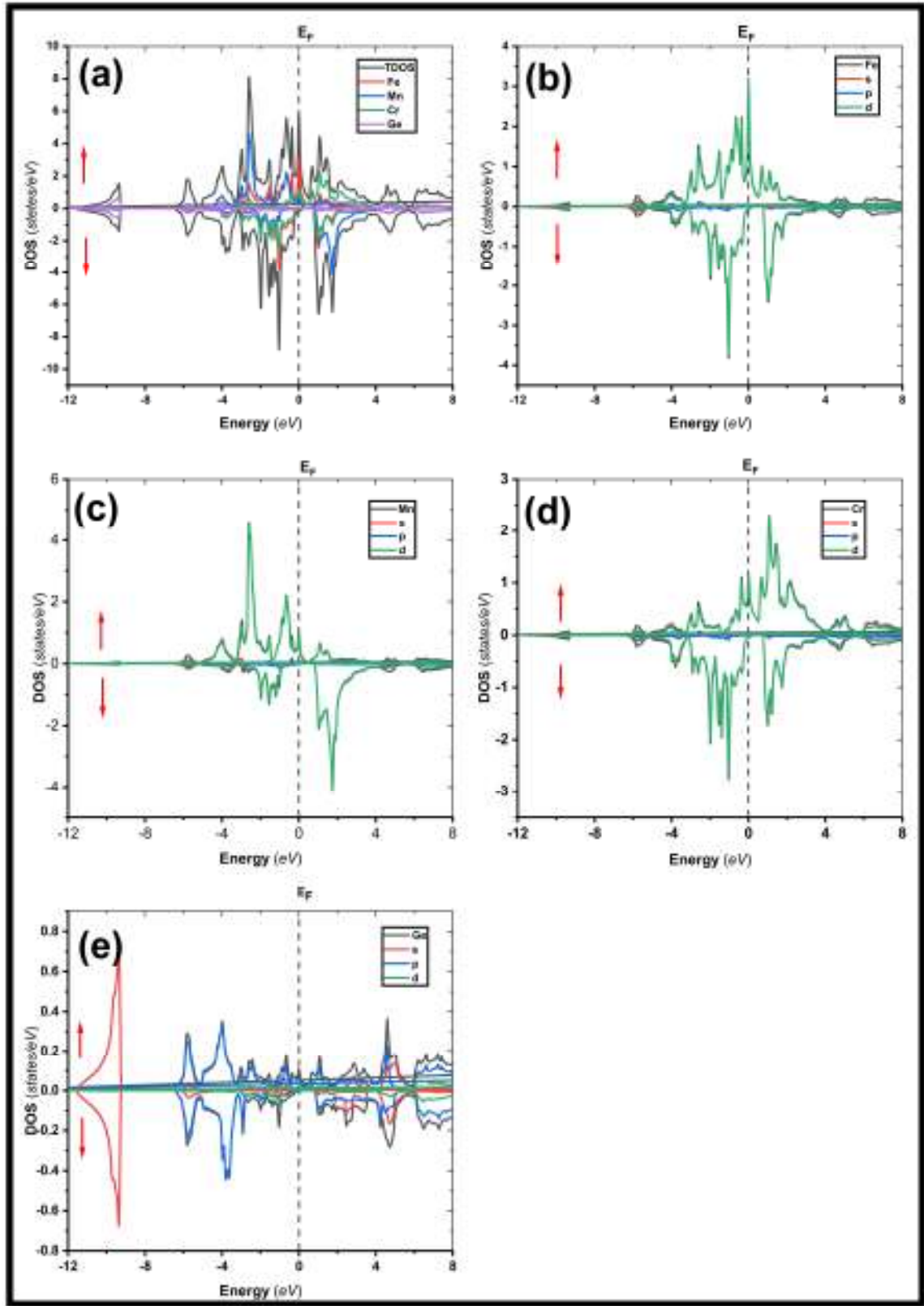
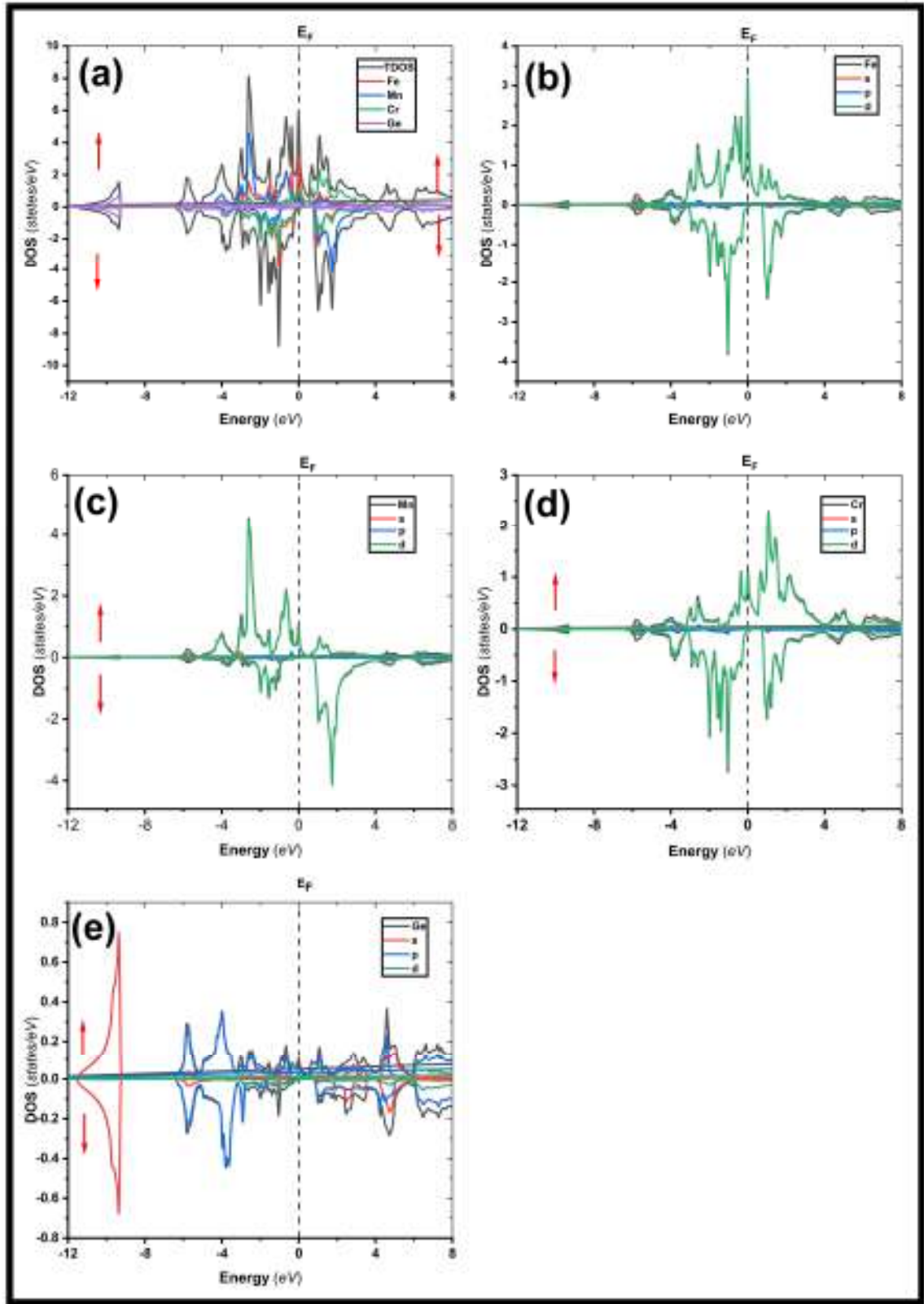


Figure 7

Total and Partial Density of States (DOS) for FeMnCrGe Using GGA Approximation



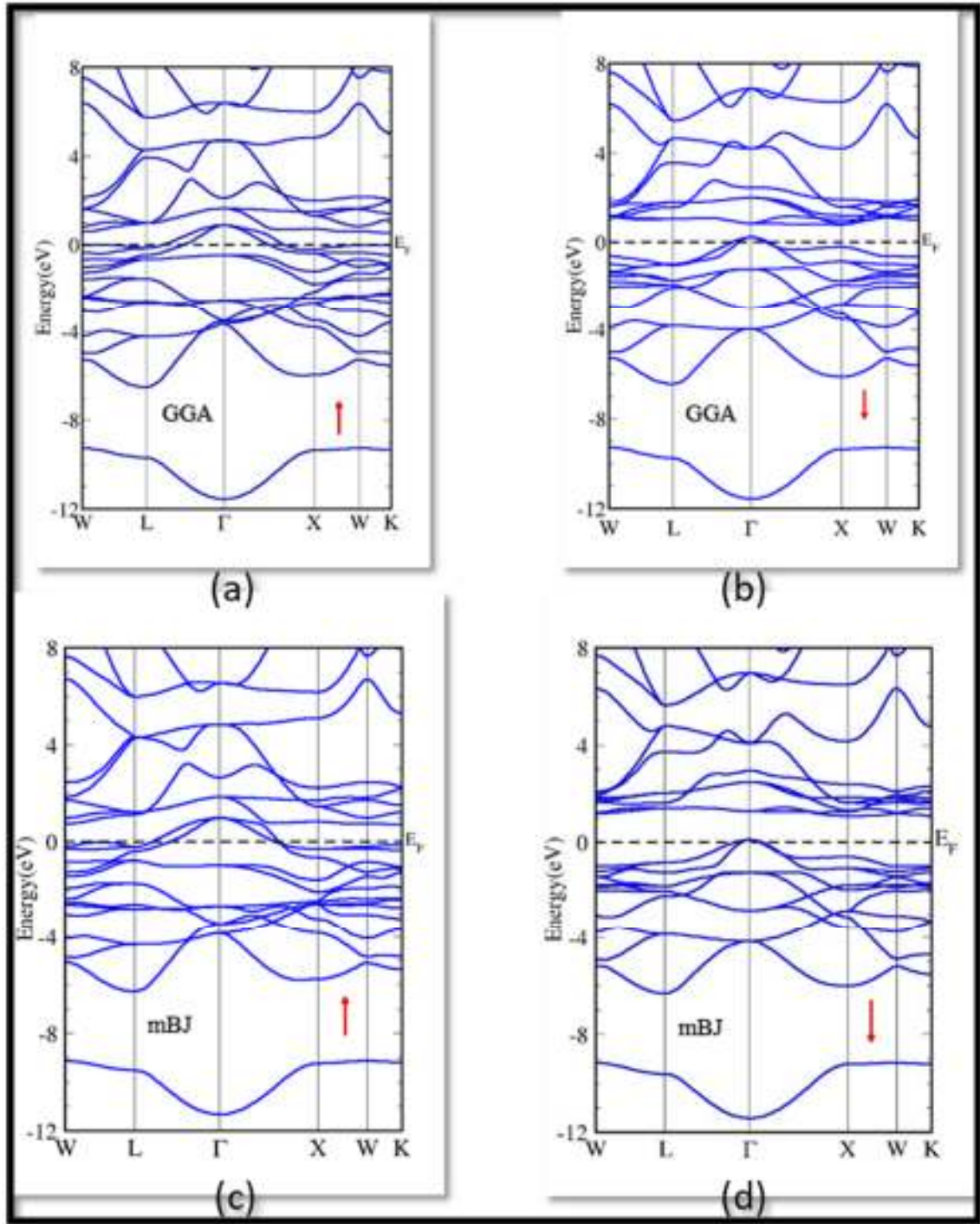
The electronic band structures of *FeMnCrGe* were examined for both spin-up and spin-down configurations, employing the Generalized Gradient Approximation and the modified Becke-Johnson potential. Figures 8 (a-d) illustrate the detailed band structures for both spin channels. While the majority spin channel exhibits metallic behavior, with bands crossing the Fermi level, the minority spin channel shows an indirect semiconductor band gap. This gap measures 0.5538 eV with GGA and 0.974 eV with mBJ.

Within the first Brillouin zone, the valence band reaches its maximum at the Γ point, while the conduction band achieves its minimum at the X point. These results unequivocally confirm the half-metallic behavior of the *FeMnCrGe* compound. Spintronic applications, particularly spin valves and magnetic tunnel junctions, demand precise electron spin manipulation for optimal performance. The semiconductor properties of the minority spin channel directly contribute to meeting this requirement, making the material highly promising.

Consequently, the *FeMnCrGe* alloy's distinctive combination of metallic behavior in the majority spin channel and semiconducting properties in the minority spin channel underscores its suitability for next-generation spintronic devices. Significant advantages for advanced applications requiring both high conductivity and spin-selective behavior are provided by this duality.

Figure 8

Band structures of FeMnCrGe: spin-up and spin-down (GGA and mBJ)

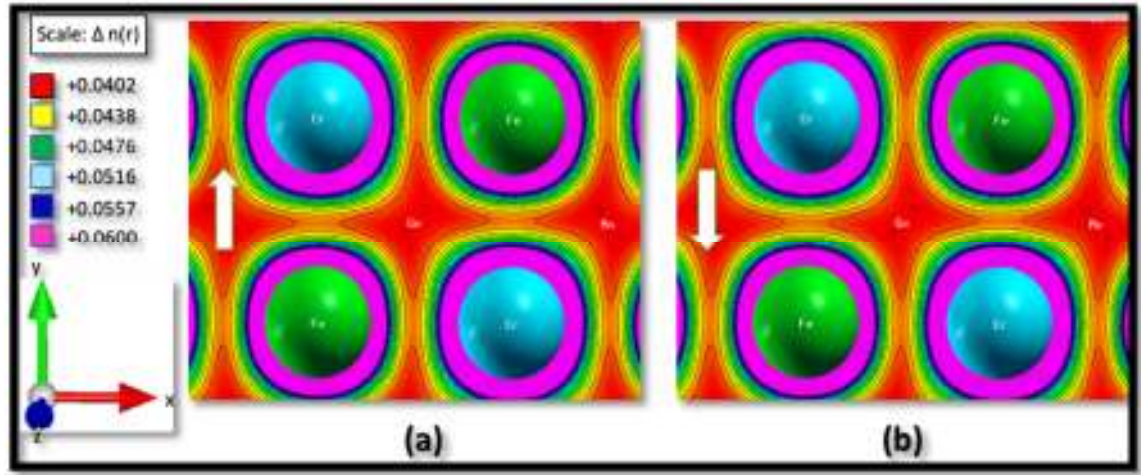


By mapping the spatial arrangement of electron density, charge density analysis provides critical insights into chemical bonding dynamics, thereby quantifying the strength and nature of atomic interactions. Such methods are foundational to modern charge distribution research. Figures 9 (a) and (b) present the two-dimensional charge density contours of the FeMnCrGe alloy for majority and minority spins within the

(110) plane. As an effective approach to visualizing chemical bonding, charge density illustrates the range of atomic interactions through electron cloud distributions. This method is extensively used in charge distribution analyses. Figures 9 (a) and (b) depict the two-dimensional charge density contours of the FeMnCrGe alloy for both majority and minority spins in the (110) plane. Ionic bonding is indicated by the spherical symmetry of the charge distribution around Fe and Mn atoms [123,124]. However, Sc and Al show elongated charge density contours that extend toward adjacent atoms, suggesting covalent bonding [125]. These bonding distinctions are most likely due to the differences in electronegativity: Fe (1.83), Mn (1.55), Cr (1.66), and Ge (2.01).

Figure 9

Two-Dimensional Charge Density Contours of FeMnCrGe in the (110) Plane for Majority and Minority Spins



3.1.5 Magnetic Properties and State Stability

The magnetic characteristics of a material are primarily defined by its total magnetic moment. For the *FeMnCrGe* alloy, interstitial, total, and partial magnetic moments were computed using the GGA and the mBJ methods, with the results provided in Table 3. According to the Slater-Pauling rule, expressed as $M_{tot} = Z_{tot} - 24$, the total magnetic moment per formula unit, M_{tot} , is determined by subtracting 24 from the sum of valence electrons, Z_{tot} . *Fe*, *Mn*, *Cr*, and *Ge* exhibit valence electron configurations of $3d^6 4s^2$, $3d^5 4s^2$, $3d^5 4s^1$, and $3d^{10} 4s^2 4p^2$, respectively. Based on these configurations, *FeMnCrGe*, with 25 valence electrons, is predicted by the rule to have a magnetic moment of $M_{tot} = 1 \mu_B$ [126]. This theoretical value is validated by calculations using both the Generalized Gradient Approximation (GGA) and the modified Becke-Johnson (mBJ)

methods, which yield a total magnetic moment of approximately $1.00 \mu B$. The magnetic moments of Mn atoms are prominently positive, recorded at $2.81240 \mu B$ with GGA and $3.10960 \mu B$ with mBJ, while Cr atoms show substantial negative moments, amounting to $-1.80556 \mu B$ under GGA and $-2.36993 \mu B$ under mBJ. Fe atoms provide smaller positive contributions, calculated as $0.10411 \mu B$ and $0.46496 \mu B$. The magnetic moments from Ge atoms are negligible, measured at $0.01965 \mu B$ for GGA and $0.02637 \mu B$ for mBJ, whereas the interstitial areas display weak negative polarization, recorded as $-0.0998 \mu B$ with GGA and $-0.22203 \mu B$ with mBJ. These findings confirm that *FeMnCrGe* complies with the Slater-Pauling rule, with a total magnetic moment close to $1.00 \mu B$, and exhibits perfect half-metallicity. The mBJ approach, which better captures d-electron localization, produces slightly higher magnetic moments for *Mn*, *Fe*, and *Cr* compared to GGA [127,128]. The alloy's ferromagnetic behavior, particularly in the transition metal regions, is further enhanced by the improved treatment of electron-electron interactions. The half-metallic nature, robust ferromagnetism, and compliance with the Slater-Pauling rule make *FeMnCrGe* an excellent candidate for spintronic applications, such as spin valves, magnetic tunnel junctions, and energy-efficient memory devices.

Table 3

Magnetic Moments of FeMnCrGe Alloy Using GGA and mBJ Methods

Property	Type 1 (GGA)	Type 1 (mBJ)	Type 2 (GGA)	Type 2 (mBJ)	Type 3 (GGA)	Type 3 (mBJ)
<i>Interstitial Moment (μB)</i>	-0.1	-0.222	0.0834	-0.159	-0.1	-0.213
<i>Fe (μB)</i>	0.1041	0.465	2.3395	2.4909	0.11	0.4535
<i>Mn (μB)</i>	2.8124	3.1096	1.6849	2.3152	2.8377	3.1274
<i>Cr (μB)</i>	-1.806	-2.37	-1.408	-2.079	-1.835	-2.385
<i>Ge (μB)</i>	0.0197	0.0264	0.0077	0.0019	0.0188	0.0258
<i>Total Magnetic Moment (μB)</i>	1.0307	1.0089	2.5411	2.5699	1.0316	1.0083

Magnetic materials display a range of ground-state behaviors that are critical for evaluating their stability and potential uses. Determining the most stable configuration among ferromagnetic (FM), paramagnetic (PM), and antiferromagnetic (AFM) states involves an analysis of total energy and structural parameters. Using the GGA-PBE approximation, the analysis identifies the FM state as the most stable.

The findings, presented in Table 4 and visualized in Figure 10, attribute this stability to the FM state's minimal total energy and superior structural properties. Figure 9 underscores the energy variations between the states, affirming the FM state's stability. Furthermore, its reduced bulk modulus implies greater structural flexibility. These results confirm that the FM state is both structurally and energetically favorable within the GGA-PBE framework.

Figure 10

Total Energy Calculations for Different Magnetic Orderings (FM, PM, AFM)

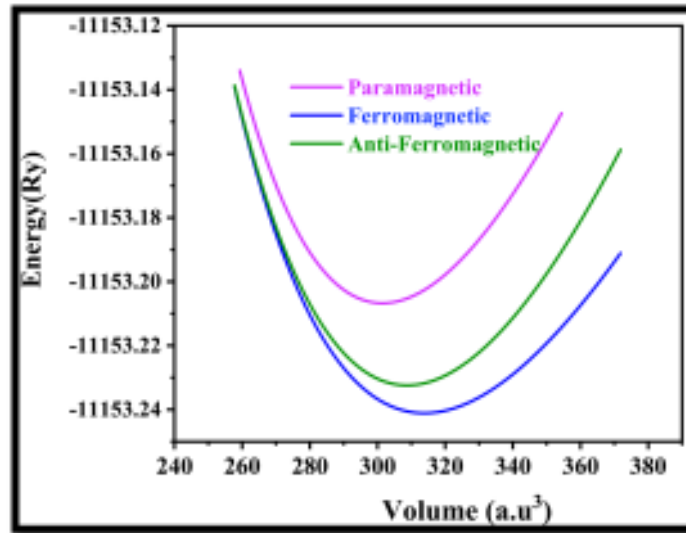


Table 4

Key parameters of ferrimagnetic, paramagnetic, and antiferromagnetic for the FeMnCrGe alloy

Property	Ferromagnetic (FM)	Paramagnetic (PM)	Antiferromagnetic (AFM)
Lattice Constant ($\text{\AA}$)	5.7085	5.6325	5.6774
Bulk Modulus (GPa)	192.91	260.19	232.53
Bulk Modulus Derivative (B')	5.3215	5.3199	4.4472
Total Energy E_{min} (Ry)	-11153.24	-11153.19	-11153.20

3.1.6 Optical properties

The quaternary Heusler alloy *FeMnCrGe* underwent a detailed optical property analysis through spin-resolved calculations. Parameters such as the dielectric function, refractive index, extinction coefficient, absorption coefficient, and energy loss were assessed over a photon energy interval of 0 to 14 eV. The Generalized Gradient Approximation

(GGA) and modified Becke-Johnson (mBJ) exchange-correlation functionals were utilized in the analysis. Figure A.1 (a-d) presents the spin-resolved dielectric function of *FeMnCrGe*, computed over the energy range of 0 to 14 eV using both GGA and mBJ. Specifically, Figure 11.A (a-b) depicts the real part, $\epsilon_1(\omega)$, and the imaginary part, $\epsilon_2(\omega)$, of the dielectric function for the majority spin channel. In materials, $\epsilon_1(\omega)$ indicates energy storage, whereas $\epsilon_2(\omega)$ reflects energy dissipation or loss [129].

As illustrated in Figure A.1 (a), the static dielectric constant $\epsilon_1(0)$ is 53.07 for both the GGA and mBJ methods. The plasma frequency (ω_p), where $\epsilon_1(\omega_p)$ equals zero, is observed at 9.18 eV for GGA and 9.48 eV for mBJ, indicating the shift between metallic and dielectric properties [130]. In Figure A.11(b), the majority spin's $\epsilon_2(\omega)$ curves show similar absorption widths for both methods, with peaks at 60.41 and 59.18, corresponding to energies of 0.39 eV and 0.36 eV, respectively. Figure A.1 (c) and (d) depict the real and imaginary parts of $\epsilon(\omega)$, for the minority spin, indicating that the peaks are primarily connected to spin-down transitions, as confirmed by the band structure and DOS analysis. As shown in Figure 11.A (c), the real part of the dielectric function provides important insights into the electronic polarizability of *FeMnCrGe*. The calculations demonstrate significant differences in the static dielectric constant, $\epsilon_1(0)$, depending on the method of computation: the GGA approach yields a value of 31.2, while the mBJ method produces a lower value of 23.6. At lower energy levels, $\epsilon_1(\omega)$ shows a significant peak near 0.8 eV for GGA and 1.21 eV for mBJ, signifying strong electronic polarization and elevated polarizability. After these peaks, a sharp decline in the dielectric function is observed, ranging from 0.8 eV to 1.2 eV for GGA and 1.21 eV to 1.46 eV for mBJ, indicating a reduction in the material's dispersive properties as photon energy increases. The real part of the dielectric function turns negative above 10.4 eV, reaching a minimum before gradually increasing toward zero at higher photon energies. This indicates a transformation in the material's optical properties, potentially influencing its electronic and wave-propagation characteristics at elevated energies.

The imaginary part of the dielectric function, $\epsilon_2(\omega)$, is shown in Figure A.1 (d), providing key insights into the material's optical absorption properties. No optical response is observed below the bandgap energy in either the GGA or mBJ approximations. However, as the photon energy exceeds the bandgap, $\epsilon_2(\omega)$ rises

sharply, reflecting the joint density of states (JDOS), which governs the available electronic transitions. Within the lower-energy region of $\epsilon_2(\omega)$, a notable peak arises, signaling substantial interband transitions in the spin-down channel [131]. These transitions are caused by incident photons, which excite electrons from the valence band to the conduction band, consistent with Equation 10 [64]. The peak is positioned around 0.93 eV for GGA and 1.36 eV for mBJ, demonstrating that the optical activity is influenced by the choice of the exchange-correlation function. As the photon energy rises, $\epsilon_2(\omega)$ declines, indicating fewer states available for interband transitions. At photon energies above 11 eV, $\epsilon_2(\omega)$ diminishes to nearly zero, indicating negligible optical absorption and emphasizing the semiconducting behavior of the spin-down state. Most optical transitions are confined to the visible and ultraviolet ranges, pointing to possible applications in UV-visible optoelectronic technologies. The variation between GGA and mBJ outcomes highlights the necessity of selecting an appropriate exchange-correlation functional for reliable optical property predictions.

Figures A.2 (a) and A.2 (c) in Appendix A present the refractive index, $n(\omega)$, and the extinction coefficient, $\kappa(\omega)$, for the majority spin channel of *FeMnCrGe*, calculated using the GGA and mBJ methods. The static values of $n(\omega)$, computed as 7.28 for GGA and mBJ, are consistent with the theoretical equation $n(0) = \sqrt{\epsilon_1(0)}$, which relates $n(\omega)$ to the real part of the dielectric function $\epsilon_1(\omega)$ [132].

The refractive index attains maximum values of 8.8 at 0.23 eV for GGA and 9.02 at 0.25 eV for mBJ, after which it gradually decreases as photon energy increases. In the ultraviolet region, *FeMnCrGe* demonstrates its lowest $n(\omega)$ values.

The extinction coefficient, $\kappa(\omega)$, illustrated in Figure A.2 (b), is closely associated with the imaginary component of the dielectric function. Figures A.2 (c) and A.12(d) illustrate the variation of $n(\omega)$ and $\kappa(\omega)$ with photon energy for the minority spin channel. In this channel, the static refractive index values, determined to be 5.58 for GGA and 4.85 for mBJ, are displayed in Figure A.2 (c). The results are in line with the theoretical relationship between $n(0)$ and $\epsilon_1(0)$ [132]. The refractive index $n(\omega)$ tracks the real part of the dielectric function, displaying notable peaks in the infrared region at 0.8 eV for GGA and 1.21 eV for mBJ. These peaks suggest the material's potential for practical applications. The refractive index in the visible spectrum maintains a relatively

high value, oscillating between 2.54 and 4.72, and progressively diminishes as photon energy rises. This pattern holds true for both computational methods. Significant optical absorption and electron transitions stimulated by photons are indicated by the sharp peaks in the extinction coefficient shown in Figure A.12(d). The maximum extinction coefficient at lower energies (around 0.9 eV for GGA and 1.3 eV for mBJ) suggests strong interband absorption. These peaks are linked to specific characteristics of the electronic band structure, which impact the material's optical properties. With increasing photon energy, the extinction coefficient decreases, suggesting a reduction in absorption efficiency.

The absorption coefficient, which measures a material's capacity to absorb photons and facilitates the resulting electronic transitions between occupied and unoccupied states [133,134], indicates how efficiently a material absorbs light at specific energies per unit thickness. The absorption coefficient for *FeMnCrGe* as a function of photon energy is presented in Figures A.3 (a) and A.3 (b), comparing the results from the GGA and mBJ exchange-correlation potentials across both spin channels. The absorption characteristics of *FeMnCrGe* in the spin-up configuration are shown in Figure A.3 (a). Consistent with its metallic nature, the alloy demonstrates substantial absorption at low photon energies. As photon energy increases, the absorption coefficient increases, peaking at 9.4 eV (GGA) and 9.83 eV (mBJ) in the ultraviolet spectrum. This behavior emphasizes the strong correlation between photon energy and absorption efficiency. The considerable absorption in the ultraviolet range is a major contributor to the alloy's relatively low extinction coefficient [135,136].

The absorption characteristics of *FeMnCrGe* in the spin-down state are presented in Figure A.13 (b). Absorption for GGA and mBJ starts at 0.55 eV and 0.97 eV, respectively, which closely match the corresponding bandgap energies predicted by these computational methods [137]. Below these energies, there is no absorption, in confirmation of the predicted lack of interaction by photons in this lower energy range. As photon energy increases, the absorption coefficient rises sharply, indicating the initiation of interband transitions at 1.12 eV (GGA) and 1.48 eV (mBJ). Attributed to electron excitation from the valence to the conduction band, the material reaches maximum absorption values of 53.4×10^4 /cm for GGA and 66.52×10^4 /cm for mBJ, reflecting its electronic structure. Several prominent peaks appear in the absorption

spectrum across the energy range, each linked to specific points in the electronic band structure where high-density states facilitate optical transitions. The absorption coefficient also demonstrates a continuous increase with photon energy over the entire spectral range. It peaks in the ultraviolet region, around 10–12 eV, indicating a strong interaction of FeMnCrGe with electromagnetic radiation in this mid- to high-energy range. Beyond 12 eV, the absorption levels off, suggesting a saturation of available states for further optical transitions. Efficient optical absorption is a challenge in optoelectronics. These results indicate that this material could solve the issue in photovoltaic devices and photodetectors. The results indicate that the material can be used in optoelectronic devices. Specifically, it is suitable for photovoltaic devices and photodetectors, which require efficient optical absorption.

Optical conductivity, symbolized as $\sigma(\omega)$, reflects the electron transitions occurring between energy levels. Its components, $\sigma_1(\omega)$ and $\sigma_2(\omega)$, are plotted against photon energy for various spin states in Appendix A, Figures A.4 (a-d). This parameter provides a quantitative measure of a material's capability to conduct electric current when exposed to electromagnetic waves.

The absorption characteristics of *FeMnCrGe* in the spin-down state are presented in Figure A.3 (b). Absorption for GGA and mBJ starts at 0.55 eV and 0.97 eV, respectively, which closely match the corresponding bandgap energies predicted by these computational methods [137]. Below these energies, there is no absorption, in confirmation of the predicted lack of interaction by photons in this lower energy range. As photon energy increases, the absorption coefficient rises sharply, indicating the initiation of interband transitions at 1.12 eV (GGA) and 1.48 eV (mBJ). Maximum absorption values of $53.4 \times 10^4 / \text{cm}$ for GGA and $66.52 \times 10^4 / \text{cm}$ for mBJ are reached, attributed to the excitation of electrons from the valence band to the conduction band, reflecting the material's electronic structure. Several prominent peaks appear in the absorption spectrum across the energy range, each linked to specific points in the electronic band structure where high-density states facilitate optical transitions. The absorption coefficient also demonstrates a continuous increase with photon energy over the entire spectral range. It peaks in the ultraviolet region, around 10–12 eV, indicating a strong interaction of *FeMnCrGe* with electromagnetic radiation in this mid- to high-energy range. Beyond 12 eV, the absorption levels off, suggesting a saturation of

available states for further optical transitions. To summarize, the absorption characteristics of *FeMnCrGe* reveal its strong capacity for photon absorption across a wide energy spectrum, with a particular emphasis on the visible and ultraviolet regions. These results underscore the material's potential for use in optoelectronic applications, including photovoltaic devices and photodetectors, where efficient optical absorption is a key requirement.

Optical conductivity, symbolized as $\sigma(\omega)$, reflects the electron transitions occurring between energy levels. Its components, $\sigma_1(\omega)$ and $\sigma_2(\omega)$, are plotted against photon energy for various spin states in Appendix A, Figures A.4 (a-d). This parameter provides a quantitative measure of a material's capability to conduct electric current when exposed to electromagnetic waves.

As illustrated in Appendix A, Figure A.4 (a), the real part of the optical conductivity, $\sigma_1(\omega)$, for the spin-up configuration of the *FeMnCrGe* alloy exhibits two key peaks. Using the Generalized Gradient Approximation (GGA), a peak of $2.7 (\Omega \cdot \text{m})^{-1}$ appears at 0.2 eV, while the modified Becke-Johnson (mBJ) method predicts a higher peak of $2.96 (\Omega \cdot \text{m})^{-1}$ at 0.4 eV. Beyond this region, additional peaks are observed at 1.62 eV and 1.86 eV for GGA, and at 2.54 eV for mBJ, followed by multiple fluctuations in the range of 3.3 eV to 7.9 eV. A pronounced peak appears at 9.7 eV (GGA) and 10.6 eV (mBJ), where the optical conductivity exceeds $8.6 (\Omega \cdot \text{m})^{-1}$. The optical conductivity shows maximum values at higher energy levels and minimal values at lower energies, indicating a dielectric-to-metallic transition that is in agreement with earlier findings.

The imaginary part of the optical conductivity, $\sigma_2(\omega)$, for the spin-up configuration, is illustrated in Figure A.4 (b). It exhibits a peak of $2.28 (\Omega \cdot \text{m})^{-1}$ at approximately 9.65 eV using GGA and $2.76 (\Omega \cdot \text{m})^{-1}$ near 9.6 eV when using mBJ. Negative values in $\sigma_2(\omega)$ are attributed to an increase in the extinction coefficient [124]. Figure A.4 (c) illustrates the real part of the optical conductivity, $\sigma_1(\omega)$, as a function of photon energy for the spin-down configuration, emphasizing the contrasts between the GGA and mBJ approximations. The minimum value of $\sigma_1(\omega)$, which aligns with the band gap energy, signifies the point of lowest conductivity. This behavior arises because optical conductivity depends on electronic transitions, which necessitate photon energies equal to or exceeding the band gap to excite electrons from the valence band to the conduction band. Two distinct peaks in the infrared spectrum of $\sigma_1(\omega)$ are seen:

one at 0.97 eV, with a conductivity of $6.77 (\Omega\cdot\text{cm})^{-1}$ for GGA, and another at 1.34 eV, reaching $9.39 (\Omega\cdot\text{cm})^{-1}$ for mBJ. The peaks reflect a substantial rise in electrical conductivity, driven by key electronic transitions in the infrared region. This is supported by the absorption coefficient data, which reveal an increase in free charge carriers due to electromagnetic radiation absorption. The agreement between optical conductivity spectra and absorption data provides strong evidence supporting the theoretical model, increasing confidence in the material's predicted infrared conductivity improvements.

Figure A.4 (d) shows the imaginary part of the optical conductivity, $\sigma_2(\omega)$, as a function of photon energy, comparing results from the GGA and mBJ approximations. Both methods display a consistent overall trend, successfully capturing the material's optical features across an extensive range of photon energies. For lower photon energies, substantial negative values of $\sigma_2(\omega)$ are observed in both GGA and mBJ results, indicating that the optical response of the material is dominated by electronic transitions at these lower energy levels. As photon energy rises, both methods exhibit peaks and fluctuations associated with interband electronic transitions. Both methods show a consistent distribution and shape of the peaks, indicating that GGA and mBJ yield comparable predictions for the material's optical behavior. This agreement underscores the validity and effectiveness of these computational techniques in modeling the optical properties of *FeMnCrGe*.

Reflectivity, $R(\omega)$, denotes the fraction of electromagnetic radiation incident on a material that is reflected at a given frequency (ω). Appendix A, Figures A.5 (a-b), show the energy-dependent reflectivity of *FeMnCrGe* for both spin states, calculated using both mBJ and GGA methods. As shown in Figure A.5 (a), the spin-up state exhibits a static reflectivity of approximately 57%, with a pronounced peak at about 66% in the infrared (IR) region for both methods. Beyond this peak, the reflectivity gradually declines, reaching a minimum in the energy range of approximately 4.3–8.8 eV. After reaching the minimum, the curves rise once more, displaying significant fluctuations before evolving into a sequence of smaller peaks and troughs, with only marginal variations. As energy increases, the reflectivity continues to decline, displaying oscillations that reflect the material's evolving interaction with the incoming radiation.

In the spin-down configuration, as shown in Figure A.5 (b), the static reflectivity, $R(0)$, at zero photon energy is calculated to be 48% using the Generalized Gradient Approximation (GGA) and 43% using the modified Becke-Johnson (mBJ) method. Both computational approaches reveal a peak reflectivity of about 66% in the infrared (IR) region. After the peak, the reflectivity curves decline steeply, with the GGA curve hitting its lowest point between 6 and 9 eV, and the mBJ curve falling to approximately 28% within the 6–10 eV range. Once these minima are reached, both curves rebound, showing pronounced fluctuations. They then transition into a sequence of smaller peaks and dips, characterized by only slight variations. Following this, both curves increase again, accompanied by sustained and noticeable oscillations.

Notably, reflectivity exhibits an inverse correlation with both conductivity and absorption across various spectral regions. The notably low reflectivity of *FeMnCrGe* establishes it as a highly promising candidate for photovoltaic applications, given that maximizing light transmission is a critical factor in optimizing the performance of optoelectronic devices. This characteristic has great advantages in enhancing the energy conversion efficiency of the device performance for technologies such as solar cells, LEDs, and photodetectors [125–127].

The energy loss function $L_{ij}(\omega)$, describes the dissipation of energy by electrons as electromagnetic waves interact with a material, shedding light on key phenomena like plasmon excitations and electronic transitions. The corresponding energy loss function $L_{ij}(\omega)$ for both spin configurations is displayed in Figures A.6 (a-b). $L_{ij}(\omega)$ steadily increases, oscillating due to electromagnetic wave interactions, as photon energy rises in the spin-up state (Figure A.6 (a)). The material's optical and electronic characteristics are significantly influenced by oscillations attributed to collective excitations, especially plasmons. After this, the curve presents peaks and fluctuations that correspond to localized electronic transitions and notable energy loss events. The steady upward trend underscores the increasing importance of higher-energy excitations and collective electron oscillations. This trend reveals a dynamic interaction between the material's electronic structure and external fields. The resulting oscillatory features are primarily caused by plasmons, collective excitations crucial for the material's optical and electronic properties. In Figure A.6 (b), corresponding to the spin-down state, the energy loss function $L_{ij}(\omega)$ shows no absorption below ~ 0.55 eV (GGA) and ~ 0.97 eV

(mBJ). This absence of significant energy dissipation in this range indicates that FeMnCrGe is minimally influenced by low-energy electromagnetic radiation, likely because of the unavailability of electronic transitions. When photon energy surpasses the defined thresholds, both curves exhibit a consistent increase in energy loss, indicating the onset of electromagnetic radiation-material interactions. This energy dissipation is primarily governed by processes like plasmonic resonances and electronic transitions. As the curves advance, minor peaks and fluctuations appear, corresponding to localized electronic transitions and distinct energy loss events. The overall upward trend in energy loss with increasing photon energy emphasizes the rising contribution of higher-energy excitations and collective electron oscillations, illustrating the complex interaction between the material's electronic structure and external electromagnetic fields. The optical properties of FeMnCrGe are distinguished by their uniqueness and specialization, as shown in our research data. These properties include a narrow band gap, significant ultraviolet absorption and reduced ultraviolet reflectivity, as well as a heightened refractive index and low reflectivity in the visible spectrum. FeMnCrGe's excellent light absorption ability makes it a prominent material for solar energy applications [15,138,139]. Moreover, its spin-dependent optical features have attracted much attention in the area of spintronics since controlling spin-polarized currents is a basic requirement for developing electronic and optoelectronic devices. The distinctive ability of this material to detect and manipulate spin-dependent optical properties renders it an excellent choice for spin-based devices such as logic circuits, spin memory, and spin sensors. Furthermore, investigating these optical signatures provides critical insights into the intricate link between magnetic and optical phenomena in advanced materials. Such study not only refines spintronic device design and functionality but also fosters a deeper comprehension of the fundamental physics. In particular, analyzing reflectivity spectra in detail unveils the material's unique properties, which can enable breakthroughs in spintronics and related technologies.

3.1.7 Thermoelectric Properties

This section explores the thermoelectric characteristics of the *FeMnCrGe* compound across a temperature span of 50 to 500 K. As shown in Figure A.7, the material's thermoelectric responses are presented as a function of temperature for both spin-up and spin-down channels in proximity to the Fermi energy level. A comparative analysis of

GGA and mBJ computational methods is presented. Figure A.7 (a) depicts the carrier concentration (N) for spin-down electrons, which starts at about 1.041 (1.017) electrons per unit cell at 50 K. As temperature increases, the concentration gradually rises, reaching about 1.042 (1.018) electrons per unit cell at 300 K and approximately 1.046 (1.02) electrons per unit cell at 500 K, consistent with findings from both methods.

For spin-up electrons, as illustrated in Figure A.7 (b), the carrier concentration (N) increases steadily with temperature. It begins at approximately -1.31 (-1.04) electrons per unit cell at 50 K, rises to about -1.056 (-1.00) electrons per unit cell at 300 K, and further reaches to nearly -1.03 (-0.97) electrons per unit cell at 500 K. These results are consistent with predictions from both GGA and mBJ methods.

As illustrated in Figure A.7 (c), the Seebeck coefficient (S) for the spin-down channel predominantly exhibits positive values, confirming the dominance of P-type charge carriers as temperature increases. According to the GGA method, S rises steadily from $42.6 \mu V K^{-1}$ at 50 K to $72.3 \mu V K^{-1}$ at 500 K, passing through $44.86 \mu V K^{-1}$ at 300 K. The mBJ method starts with $-35.5 \mu V K^{-1}$ at 50 K, transitions to positive values around 100 K, and increases to $86.04 \mu V K^{-1}$ at 300 K and $124.1 \mu V K^{-1}$ at 500 K. The findings offer further evidence supporting the dominance of P-type charge carriers at higher temperatures.

The Seebeck coefficient for the spin-up channel is illustrated in Figure A.7 (d). According to the GGA method, the coefficient starts at approximately $-15.65 \mu V K^{-1}$ at 50 K, increases to $-2.62 \mu V K^{-1}$ at 300 K, and reaches $-2.14 \mu V K^{-1}$ at 500 K. In contrast, the mBJ method shows an initial value of $5.16 \mu V K^{-1}$ at 50 K, which decreases to $-2.11 \mu V K^{-1}$ at 100 K before rising to approximately $2.6 \mu V K^{-1}$ at 500 K. The Seebeck coefficient for the spin-up channel displays both positive and negative values, mainly negative, indicating the prevalence of n-type charge carriers at elevated temperatures. This spin channel demonstrates metallic behavior, as evidenced by the smaller absolute Seebeck coefficient values relative to the spin-down channel, which exhibits semiconducting properties [140,141]. At room temperature, Seebeck coefficients ranging from 50 to $100 \mu V K^{-1}$ are generally regarded as moderately high, highlighting the *FeMnCrGe* alloy's potential for thermoelectric applications [142].

Figure A.7 (e) illustrates the electrical conductivity of the *FeMnCrGe* compound in the spin-down channel. At 50 K, the conductivity is roughly $3.18 \times 10^{19} (\Omega \text{ m s})^{-1}$ for the GGA approach, while the mBJ method yields a lower value of $1.43 \times 10^{19} (\Omega \text{ m s})^{-1}$. At 300 K, the conductivity decreases to $3.08 \times 10^{19} (\Omega \text{ m s})^{-1}$ with the GGA method and $1.36 \times 10^{19} (\Omega \text{ m s})^{-1}$ with mBJ, while at 500 K, a marginal rise brings the values to $3.15 \times 10^{19} (\Omega \text{ m s})^{-1}$ for GGA and $1.52 \times 10^{19} (\Omega \text{ m s})^{-1}$ for mBJ.

As illustrated in Figure A.7 (f), the conductivity of the spin-up channel increases with temperature, starting near $2.11 \times 10^{20} (\Omega \text{ m s})^{-1}$ at 50 K, peaking at $2.63 \times 10^{20} (\Omega \text{ m s})^{-1}$ at 300 K, and stabilizing close to $2.30 \times 10^{20} (\Omega \text{ m s})^{-1}$ by 500 K. According to the mBJ method, it begins at $3.37 \times 10^{20} (\Omega \text{ m s})^{-1}$ at 50 K, dips to its lowest value of $3.23 \times 10^{20} (\Omega \text{ m s})^{-1}$ near 100 K, then rises to $3.31 \times 10^{20} (\Omega \text{ m s})^{-1}$ at 300 K before falling slightly to $1.6 \times 10^{20} (\Omega \text{ m s})^{-1}$ at 500 K. This study reveals the contrasting behavior of electrical conductivity in the spin-up and spin-down channels of *FeMnCrGe*, highlighting differences in their electronic characteristics. The notably higher conductivity in the spin-up channel near the Fermi level strongly indicates the compound's half-metallic properties.

The electronic contribution to thermal conductivity (κ_e) was assessed for both spin channels by employing the GGA approximation and the mBJ exchange potentials. As demonstrated in Figures A.7 (g), the data support this interpretation. The electron thermal conductivity (κ_e) varies nearly linearly with temperature for both spin channels. The spin-up channel, however, displays significantly greater κ_e values than the spin-down channel, reaffirming *FeMnCrGe*'s half-metallic nature. Such insights are critical for understanding thermal transport in spintronic materials.

In Figures A.8 (a-b), the calculated thermoelectric Figure of merit (ZT) for the *FeMnCrGe* alloy under varying temperature conditions for both spin channels is illustrated using the GGA and mBJ approaches. For spin-down states, Figure A.8 (a) shows that under GGA at 50 K, the ZT value is 7.77×10^{-2} . With increasing temperature, the ZT value decreases initially, reaching 4.3×10^{-2} at 100 K and declining further to 3.7×10^{-2} by 150 K. By 300 K, however, the ZT rises to 7.9×10^{-2} and shows further improvement, culminating at 18×10^{-2} at 500 K. When analyzed using the mBJ method, the spin-down ZT begins at 6×10^{-2} at 50 K, dips sharply to 4.8×10^{-3} at 100 K, and then increases significantly, achieving 0.24 at 300 K and 0.42 at 500 K.

For spin-up states, as depicted in Figure A.8 (b), the GGA method yields a ZT of 9.09×10^{-3} at 50 K, which decreases sharply to 2.41×10^{-3} at 100 K and further diminishes to approximately 2.7×10^{-4} by 500 K. The mBJ method for spin-up states starts with a ZT of 1.21×10^{-4} at 50 K, reduces slightly to 1.81×10^{-4} at 100 K, and stabilizes around 1.2×10^{-4} at higher temperatures. At room temperature, the ZT values for the *FeMnCrGe* alloy are calculated as 3.02×10^{-4} using the GGA approach and 5.24×10^{-5} using the mBJ method. These thermoelectric Figure of merit results underscore the alloy's promising potential for thermoelectric applications, particularly in high-temperature spin-down states. The notable increase in ZT with the mBJ approach, from 0.06 at low temperatures to 0.42 at 500 K, underscores the crucial role of charge carriers, such as electrons and holes, in enhancing electrical conductivity at higher temperatures. This enhancement highlights the alloy's suitability for high-temperature energy conversion and its effectiveness in converting heat into electricity.

Material thermoelectric performance relies critically on the power factor ($PF = S^2\sigma$). Figures A.9 (a-b) illustrate how the power factor varies with temperature in both spin channels for the *FeMnCrGe* compound. Specifically, Figure A.19 (a) shows the spin-down channel, where GGA calculations yield a PF of 5.7×10^{10} at 50 K. This value decreases to 3.1×10^{10} at 100 K, and further to 2.78×10^{10} at 150 K, before rising again to 6.2×10^{10} at 300 K. From there, it continues to rise gradually, reaching 1.6×10^{11} at 500 K.

Figure A.9 (b) indicates that for the spin-up channel, the PF at 50 K is 9.09×10^{-3} for GGA and 1.21×10^{-3} for mBJ. As the temperature rises, the PF decreases notably, dropping to 2.92×10^{-4} for GGA and 1.32×10^{-4} for mBJ at 300 K. The PF further decreases to 2.04×10^{-4} for GGA and 2.97×10^{-4} for mBJ by 500 K.

The power factor PF values show a clear temperature dependence, particularly in the spin-down state of *FeMnCrGe*. At 500 K, the power factor PF peaks at approximately 1.6×10^{11} , indicating the alloy's improved thermoelectric performance and exceptional electrical conductivity at high temperatures. The increase in PF is directly connected to the rise in ZT for spin-down states, revealing that *FeMnCrGe* achieves its highest thermoelectric efficiency at high temperatures. The mBJ method further enhances this improvement by precisely modeling electronic interactions and improving charge

carrier mobility, highlighting the alloy's strong suitability for thermoelectric applications under high-temperature conditions.

Illustrated in Figures A.10 (a-b), the Hall coefficient (R_H) demonstrates a clear temperature dependence. Figure A.10 (a) displays the R_H values for the spin-down channel, calculated using both the GGA and mBJ methods. The R_H for GGA, measured at $6.85 \times 10^{-9} \text{ m}^3/\text{C}$ at 50 K, exhibits stable behavior with only minor temperature fluctuations. While the R_H follows a minor fluctuation between 300 K and 500 K, increasing slightly before decreasing, the mBJ method demonstrates a more pronounced variation. It begins at a significantly higher value of $1.82 \times 10^{-8} \text{ m}^3/\text{C}$ at 50 K, peaks at $1.92 \times 10^{-8} \text{ m}^3/\text{C}$ at 100 K, and then gradually declines to $1.45 \times 10^{-8} \text{ m}^3/\text{C}$ at 500 K.

For spin-up states, the Hall coefficient R_H shows consistent stability across the 50-500 K temperature range for both approximation methods. As depicted in Figure A.10 (b) in Appendix A, the R_H for GGA starts at $1.82 \times 10^{-10} \text{ m}^3/\text{C}$ at 50 K, decreases to $1.56 \times 10^{-10} \text{ m}^3/\text{C}$ at 300 K, and then increases to $1.72 \times 10^{-10} \text{ m}^3/\text{C}$ at 500 K. Starting at $1.10 \times 10^{-10} \text{ m}^3/\text{C}$ at 50 K, the R_H for mBJ decreases slightly to $1.04 \times 10^{-10} \text{ m}^3/\text{C}$ at 300 K but eventually rises back to its initial value at 500 K. The positive R_H sign confirms the presence of hole carriers, characteristic of P-type conduction [143].

These findings demonstrate distinct characteristics for each spin channel, emphasizing the compound's half-metallic nature and potential use in high-temperature thermoelectric devices. The higher ZT and PF values of the spin-down channel indicate its superior thermoelectric performance, while the spin-up channel exhibits typical metallic behavior with lower Seebeck coefficients. By more accurately modeling electronic correlations, the mBJ approximation method consistently improved the results, emphasizing the *FeMnCrGe* compound's suitability for spintronics and thermoelectric applications at elevated temperatures.

The investigation using the Slack model reveals that the thermal transport properties of the FeMnCrGe alloy vary systematically with temperature [100,101]. Heat conduction via atomic vibrations peaks at 1.81 at 50 K, then decreases steadily to 0.18 at 500 K, with the rate of decline slowing at higher temperatures, as shown in Figure A.11. In both GGA and mBJ analyses, the total thermal conductivity (κ_{Tot}) for the two spin channels, shown in Figures A.12 (a) and (b), follows a pattern akin to electronic thermal

conductivity ($\square e$), growing with temperature. This suggests that lattice thermal conductivity has a minimal impact.

3.2 FeMnScAl Compound

3.2.1 Structural and Thermodynamic Stability of *FeMnScAl*

The *FeMnScAl* compound adopts a face-centered cubic structure, crystallizing in the Fm-43 space group (number 216) and following the *LiMgPdSn*-type configuration. To determine the most stable atomic arrangement, three configurations were examined, taking into account the cubic structure's symmetry and atomic occupancy positions. These structural configurations were analyzed using a unique volume optimization technique. The investigation was guided by the Murnaghan equation of state, which mathematically represents total energy and pressure as functions of volume per formula unit, as depicted in Equation (57). With the Al atom from the main group located at the 4d site (0.75, 0.75, 0.75), three distinct configurations are possible for arranging Fe, Mn, and Sc atoms among the 4a (0, 0, 0), 4c (0.25, 0.25, 0.25), and 4b (0.5, 0.5, 0.5) sites, as outlined in Table 1 [107–109]. These structural arrangements were examined using a volume optimization approach, guided by the Murnaghan equation of state, which models total energy and pressure as a function of volume per formula unit, as shown in Equation (57) [28,110]. The energy-volume relationship for the three atomic arrangements of *FeMnScAl* is shown in Appendix A, Figure A.13. The most stable configuration, *FeMnScAl* type 2, positions Fe at the 4a site (0, 0, 0), Mn at the 4b site (0.5, 0.5, 0.5), Sc at the 4c site (0.25, 0.25, 0.25), and Al at the 4d site (0.75, 0.75, 0.75). When comparing energy values at zero pressure and absolute zero temperature, this arrangement exhibits the lowest energy.

Table 5 The key equilibrium structural parameters, including optimized energy (E_0), lattice parameter (a_0), bulk modulus (B), and its derivative (B'), are compiled in Table 5.

Table 5*Atomic configuration and structural properties of FeMnScAl.*

Property/Atom	Structure Type 1	Structure Type 2	Structure Type 3
Fe Atom Position	(0,0,0)	(0,0,0)	(1/4,1/4,1/4)
Mn Atom Position	(1/4,1/4,1/4)	(1/2,1/2,1/2)	(0,0,0)
Sc Atom Position	(1/2,1/2,1/2)	(1/4,1/4,1/4)	(1/2,1/2,1/2)
Al Atom Position	(3/4,3/4,3/4)	(3/4,3/4,3/4)	(3/4,3/4,3/4)
Lattice Constant a (Å)	6.1532	6.1142 6.2 [144]	6.215
Total Energy E_0 (Ry)	-6877.00151	-6877.0524	-6877.01941
Bulk Modulus B (GPa)	106.63	119.57	96.3299
Bulk Modulus (B')	5.5122	5.0653	4.2207
Formation Energy E_{Form} (Ry)	-0.6816	-0.7325	-0.6995
Cohesive Energy (Ry/Atom)	1.236042	1.286932	1.253942

Figure A.14 provides a visual representation of the crystalline arrangement of the *FeMnScAl* compound, illustrating three distinct structural types and the associated Brillouin zone, including high-symmetry points and reciprocal vectors.

The thermodynamic stability of the *FeMnScAl* compound is determined by its formation energy E_{Form} [110,111], which measures the energy change during the compound's synthesis from its elements under standard conditions [112]. Equation 55 was used to calculate the formation energy E_{Form} for the three atomic configurations of *FeMnScAl* [29]:

$$E_{Form} = E_{FeMnScAl(x)}^{Cubic} - (E_{Fe}^{Cubic} + E_{Mn}^{Cubic} + E_{Sc}^{Cubic} + E_{Al}^{Cubic}) \dots\dots\dots (58)$$

Here, $E_{FeMnScAl(x)}^{Cubic}$ represents the compound's lowest energy, while E_{Fe}^{Cubic} , E_{Mn}^{Cubic} , E_{Sc}^{Cubic} and E_{Al}^{Cubic} denote the minimum energies of *Fe*, *Mn*, *Sc*, and *Al*, respectively, with ($x = 1, 2, 3$) indicating the configuration type. The negative formation energy values at zero Kelvin and zero pressure, as presented in Table 1, suggest the alloy's stability.

The cohesive energy of FeMnScAl in its three structural variations is calculated by subtracting the total energy of the compound from the sum of its isolated atomic energies and normalizing by the number of atoms in the unit cell [114].

$$E_{coh.} = \frac{(E_{Fe}^{iso} + E_{Mn}^{iso} + E_{Sc}^{iso} + E_{Al}^{iso}) - E_{FeMnScAl(x)}^{Cubic}}{4} \dots\dots\dots (59)$$

A key indicator of structural stability, this parameter demonstrates favorable configurations through positive values. Table 5 reports the cohesive energy values, which, when examined together with mechanical stability findings, establish the compound's potential for extended research and practical applications.

3.2.2 Phonon Analysis of FeMnScAl's Dynamical Stability

The dynamic stability of the compound under investigation was evaluated by computing its phonon dispersion curve using the linear response method available in the CASTEP code [145]. Figure A.15 displays the resulting phonon dispersion diagram. If a material's phonon spectrum does not exhibit soft modes, which are imaginary modes characterized by negative frequencies, it is deemed dynamically stable. The appearance of soft modes frequently suggests lattice instability, which may result in structural phase transitions that complicate synthesis and restrict potential applications. As shown in Figure A.15, FeMnScAl does not display any soft modes, confirming its dynamic stability [122,146–149]. These findings confirm the robustness of the lattice structure, demonstrating its capacity to withstand deformation and resist phase transitions under standard operating conditions. This durability enhances fabrication efficiency while facilitating its incorporation into advanced technological developments.

3.2.3 Charge Density and Bond Analysis

As a powerful tool for depicting chemical bonding, charge density reveals the interaction range between atoms through the distribution of electron clouds. This method is commonly utilized in studies of charge distribution. The two-dimensional charge density contours of the *FeMnScAl* alloy for both majority and minority spins are shown in Figures A.16 (a) and (b) within the (110) plane. Ionic bonding is indicated by the spherical symmetry of the charge distribution around Fe and Mn atoms [123,124]. In contrast, the charge density contours around Sc and Al, which are elongated and directed toward neighboring atoms, reveal covalent interactions with these atoms [125].

The variation in bonding nature is likely influenced by the differing electronegativities of the elements: Fe (1.83), Mn (1.55), Sc (1.36), and Al (1.61).

3.2.4 Elastic Properties

The material's mechanical stability is confirmed when its elastic constants fulfill the following conditions: $C_{11} > 0$, $C_{11} + 2C_{12} > 0$, $C_{12} < B < C_{11}$, $C_{11} - C_{12} > 0$, and $C_{44} > 0$. The findings demonstrate that *FeMnScAl* complies with these requirements, establishing its mechanical stability [119]. Under conditions of zero strain, zero pressure, and absolute zero temperature, the mechanical properties of FeMnScAl were calculated and are summarized in Table 6.

Table 6

Mechanical Properties of FeMnScAl Alloy Configurations at zero strain ($\delta = 0$)

Property	Type 1	Type 2	Type 3	Units
Elastic Constant C_{11}	111.573	154.9892	85.624	GPa
Elastic Constant C_{12}	102.778	103.6927	69.996	GPa
Elastic Constant C_{44}	93.508	75.6698	79.593	GPa
Shear Modulus (S_R)	10.269	42.508	17.027	GPa
Bulk Modulus (B)	105.71	120.791	75.205	GPa
B/S_R Ratio	10.29409	2.841606	4.416809	-
Young's Modulus (Y)	29.84	114.135	47.496	GPa
Poisson's Ratio (ν)	0.452	0.342	0.394	-
Anisotropic Factor (A)	21.2639	2.950291	10.18594	-

This study investigated the mechanical stability and chemical bonding of the QHA *FeMnScAl*. The calculated shear and bulk moduli highlight the material's ability to withstand deformation under applied stress [37]. The shear modulus (S_R) and bulk modulus (B) further illustrate the material's resistance to stress-induced deformation. A high Young's modulus indicates stiffness, while a low value suggests greater flexibility [113]. Poisson's ratio is a crucial parameter for evaluating a material's response to stress, stability, and bonding behavior. For classification, a ratio above 0.26 signifies ductility, while values below this threshold indicate brittleness. Since all three FeMnScAl types possess Poisson's ratios exceeding 0.26, they are classified as ductile [150,151]. FeMnScAl's B/S_R values, which exceed the brittleness threshold of 1.75 by a

wide margin, confirm its classification as a ductile material [120]. Poisson's ratios less than 0.25 are indicative of ionic bonding, whereas values greater than 0.25 correlate with covalent bonding. For FeMnScAl , the Poisson's ratios correspond to covalent bonding [44]. In addition, the anisotropic factor (A), diverging from unity, demonstrates the material's mechanical anisotropy [37,47]. These conclusions are significant for predicting its behavior in industrial applications.

The electronic, optical, and thermoelectric analyses and results will focus exclusively on the Type 2 structure. This structure has been identified as both thermodynamically, dynamically, and mechanically stable, and it possesses the lowest energy configuration.

3.2.5 Electronic Properties

Understanding chemical bonding in elements and their alloys requires a thorough study of electronic properties, which sheds light on the fundamental interactions between constituent elements. In this work, the density of states (DOS) and band structure of the FeMnScAl alloy (type 2) are examined. Figures A.17 (a-e) and A28 (a-e), analyzed using the GGA and mBJ approximations, show the total and partial DOS, offering a detailed view of the energy band gap's origin. Both methods indicate that the DOS close to the Fermi level is primarily shaped by the d-orbitals of Fe , Cr , and Sc in both the conduction and valence bands, with minimal contributions from p - and s -orbitals in both spin channels. The compound's half-metallic nature, characterized by 100% spin polarization due to the presence of spin-up states and the absence of spin-down states at the Fermi level, underscores its potential for spintronic applications.

Depicted in Figure A.19 (a-d), the electronic band structure of the FeMnScAl alloy was analyzed for spin-up and spin-down states via GGA and mBJ methods. These results validate the metallic nature of the majority spin channel, indicated by bands crossing the Fermi level, while the minority spin channel shows semiconducting features with an indirect bandgap. The size of the gap varies depending on the method used: 0.14562 eV for GGA and 0.1574 eV for mBJ, illustrating the influence of the exchange-correlation potential treatment. By considering the electron density gradient, GGA provides useful approximations but frequently underestimates the potential in zones with sharp density variations, resulting in smaller bandgap values. Alternatively, mBJ employs a semi-local framework that incorporates both density and kinetic energy density, yielding a

more accurate portrayal of exchange-correlation effects near the Fermi level. Thus, mBJ's calculations demonstrate improved alignment with experimental data, fostering a more comprehensive understanding of electronic properties [106]. The enhanced precision of mBJ over GGA makes it a critical asset in material prediction and screening [94]. In the first Brillouin zone, the valence band achieves its maximum at the X point, while the conduction band drops to its minimum at the L point, affirming the half-metallic behavior of the *FeMnScAl* alloy.

3.2.6 Magnetic Properties

A material's magnetic behavior is principally defined by its total magnetic moment. For the *FeMnScAl* alloy, the interstitial, total, and partial magnetic moments were calculated via the GGA and mBJ methods, as shown in Table 7. The type 2 version of this alloy follows the Slater-Pauling rule, $M_{\text{tot}} = Z_{\text{tot}} - 24$, where M_{tot} is the total magnetic moment per formula unit and Z_{tot} is the total number of valence electrons. The valence configurations for the elements are *Fe* ($3d^6 4s^2$), *Mn* ($3d^5 4s^2$), *Sc* ($3d^1 4s^2$), and *Al* ($3s^2 3p^1$). According to the Slater-Pauling rule, a total magnetic moment of 3 μ_B is predicted for the alloy's 21 valence electrons, which closely matches the calculated magnetic moments for both type 1 and type 2 *FeMnScAl* alloys.

Analysis of the magnetic moment distribution shows strong positive contributions from Mn atoms, notable negative contributions from Cr atoms, modest positive contributions from Fe atoms, and negligible contributions from Ge atoms. The *FeMnScAl* alloy exhibits a total magnetic moment of 3.00 μ_B , underscoring its ideal half-metallic nature. The total magnetic moment for type 3 is also non-integer. These findings are consistent with the research conducted by Y. C. Gao and X. Gao on the magnetic behavior of the *FeMnScAl* alloy [144]. Optical and thermoelectric analyses will center exclusively on the type 2 structure, which has been determined to be both thermodynamically and mechanically stable. Additionally, this structure exhibits the most pronounced half-metallic band gap, highlighting its distinctive properties.

Table 7*Magnetic properties of FeMnScAl alloy: Type 1, Type 2, and Type 3 Structures*

Parameter	Type 1	Type 1	Type 2	Type 2	Type 3	Type 3
Method	GGA	mBJ	GGA	mBJ	GGA	mBJ
Interstitial	0.00845	-0.19137	-0.07580	-0.03687	-0.22531	-0.3510
μ_{Fe}	0.55277	0.63505	0.42708	0.02084	1.83863	1.95022
μ_{Mn}	2.52363	2.71301	2.85759	3.15503	2.85152	3.05055
μ_{Sc}	-0.04105	-0.09468	-0.15056	-0.09040	-0.25582	-0.3180
μ_{Al}	-0.04392	-0.06201	-0.05880	-0.04886	-0.09485	-0.1284
μ_{Tot}	2.99987	3.00000	2.99951	2.99973	4.11416	4.20329

FeMnScAl exhibits distinct structural and energetic differences based on its magnetic configuration, which can be antiferromagnetic (AFM), ferromagnetic (FM), or paramagnetic (PM). The AFM and FM phases share nearly identical cubic lattice parameters of about 6.116 Å, whereas the PM state has a smaller lattice parameter of 6.0297 Å, indicating increased atomic density. These differences are depicted in Figure A.20 and detailed in Table 8.

A bulk modulus of 142.86 GPa makes the PM state the most resistant to compression. In contrast, the FM state, which has a bulk modulus of 119.57 GPa, is the least resistant to compressive stress. The pressure derivative of the bulk modulus also sheds light on these differences: the AFM state achieves the highest value of 5.1484, implying a unique stiffening mechanism under pressure that sets it apart from the FM (5.0653) and PM (4.3020).

The FM state's equilibrium energy of -6877.089042 Ry marks it as the most stable, while the PM state's energy of -6877.029317 Ry suggests lower stability. These findings illustrate the connection between magnetic ordering, structural density, and mechanical resilience, with the FM state identified as the thermodynamically preferred state under equilibrium. This information provides key insights for enhancing material characteristics in applications such as magnetic storage and pressure-responsive technologies.

Table 8*Magnetic Phase-Dependent Properties of FeMnScAl (AFM, FM, and PM)*

Property	Anti-Ferromagnetic	Ferromagnetic	Paramagnetic
Cubic Lattice Parameter ($\text{\AA}$)	6.1168	6.1167	6.0297
$V_0 (a.u.)^3$	385.0216	386.1023	369.7902
B (GPa)	120.8335	119.57	142.8609
B'	5.1484	5.0653	4.3020
E_0 (Ry)	-6877.051895	-6877.089042	-6877.029317

3.2.7 Optical properties

Illustrated in Figure A.21 (a-d), the dielectric characteristics of *FeMnScAl* are investigated over a wide energy interval from 0 to 8 eV, covering the infrared (IR), visible light (VL), and ultraviolet (UV) spectra. For the spin-up configuration, detailed in Figure A.21 (a), the static dielectric constant $\epsilon_I(0)$ is found to be 217.67 with the GGA method and 129.13 using the mBJ technique at the lower limit of $\epsilon_I(\omega)$. As a key parameter, the static dielectric constant measures a material's capacity to neutralize electric fields produced by localized charges, reflecting its response to low-frequency or static electric conditions [152]. Notably, the first plasma frequency (ω_p), where $\epsilon_I(\omega_p)$ reaches zero, is determined to be 0.286 eV for GGA and 0.422 eV for mBJ. At this frequency, the material shifts from metallic to dielectric behavior [120]. Pronounced peaks in the dielectric function, calculated using GGA and mBJ methods, are observed in the infrared spectrum for the *FeMnScAl* alloy. Additionally, negative $\epsilon_I(\omega)$ values indicate metallic characteristics in specific infrared and ultraviolet regions [153], highlighting the alloy's distinctive electronic properties.

This material exhibits considerable promise for use in optical filters or shielding devices that function over a broad spectrum of frequencies. As illustrated in Figure A.21 (b), the $\epsilon_2(\omega)$ curves for the majority spin, indicating the absorption region's breadth, reveal a strong alignment between the GGA and mBJ methods. The peak values of $\epsilon_2(\omega)$ are located at 114.98 and 129 for GGA and mBJ, respectively, corresponding to energies of 0.28 eV and 0.31 eV. Starting from zero eV, the $\epsilon_2(\omega)$ values confirm the absence of an optical band gap, further supporting the material's metallic nature in this state.

In Figure A.21 (c), the $\epsilon_1(\omega)$ curve for the spin-down state reveals a static dielectric constant $\epsilon_1(0)$ of 120 for GGA and 129.13 for mBJ. Optical transitions are characterized by peaks at 0.14562 eV in the GGA results and 0.1574 eV in the mBJ results. These peaks, which decrease sharply in the infrared region, result from inter-band transitions of bonding electrons. Such behavior highlights the intense interactions between energy bands in this spectral range.

As depicted in Figure A.21 (d), the imaginary part of the dielectric function, $\epsilon_2(\omega)$, indicates the material's ability to absorb radiation. Peaks in $\epsilon_2(\omega)$ occur at 0.45 eV for the GGA method and 0.37 eV for the mBJ approach. Peaks in $\epsilon_2(\omega)$ are observed at 0.45 eV for the GGA method and 0.37 eV for the mBJ approach. Significantly, $\epsilon_2(\omega)$ stays at zero until the photon energy surpasses the band gap, marking the onset of absorption and the initiation of direct optical transitions from the valence band maximum to the conduction band minimum.

Figure A.22 (a-d) displays the refractive index, $n(\omega)$, and the extinction coefficient, $\kappa(\omega)$, for the *FeMnScAl* alloy across both majority and minority spin channels. Figures A.22 (a) and (b) illustrate the majority spin state, while Figures A.22 (c) and (d) show the minority spin state, computed using GGA and mBJ methods. For the majority spin channel, the static refractive indices, determined as $n(0) = \sqrt{\epsilon_1(0)}$, are found to be 11.36 (GGA) and 14.73 (mBJ), consistent with the relationship between $n(\omega)$ and the real part of the dielectric function, $\epsilon_1(\omega)$ [132]. Peaks in $n(\omega)$ are observed at 14.8 (GGA) at 0.05 eV and 11.3 (mBJ) at 0.23 eV, after which they decrease as photon energy rises. In the ultraviolet spectrum, *FeMnScAl* exhibits its lowest refractive index values. The extinction coefficient, $\kappa(\omega)$, shown in Figure A.22 (b), closely aligns with the imaginary part of the dielectric function, highlighting their intrinsic relationship. For the minority spin channel, Figures A.22 (c) and A.32 (d) depict the relationship between photon energy and the refractive index, $n(\omega)$, as well as the extinction coefficient, $\kappa(\omega)$. As illustrated in Figure A.22 (c), the static values of $n(\omega)$ are 10.8 for GGA and 11.26 for mBJ, consistent with the theoretical relationship between $n(0)$ and $\epsilon_1(\omega)$ [132]. Similar to $\epsilon_1(\omega)$, the refractive index exhibits peaks in the infrared region at approximately 0.2 eV for GGA and 0.28 eV for mBJ. High $n(\omega)$ values, ranging from 2.9 to 3.93, persist in the visible spectrum but gradually decrease with rising photon energy, with both methods yielding comparable results. In Figure A.22 (d), the

extinction coefficient $k(\omega)$ closely tracks $\epsilon_2(\omega)$, staying at zero until photon energy exceeds the band gap, at which point absorption begins. The coefficient exhibits distinct peaks, linked to intense optical absorption and transitions driven by photon energy. Both GGA and mBJ methods show notable peaks near 0.64 eV at lower energy ranges, which correspond to strong absorption.

A detailed investigation of the optical absorption properties of *FeMnScAl* is shown in Figures A.23 (a) and A.33 (b), utilizing GGA and mBJ exchange-correlation potentials for both majority and minority spin channels. In the spin-up channel (Figure A.19 (a)), the absorption reveals metallic behavior, starting at lower photon energies and increasing gradually as the photon energy grows. Within the ultraviolet range, the absorption attains its maximum value at roughly $\sim 196 \times 10^4/cm$ for both GGA and mBJ, consistent with the material's low extinction coefficient, as noted in references [139,154].

The spin-down channel in Figure A.23 (b) shows distinct absorption thresholds at 0.145 eV for GGA and 0.157 eV for mBJ, corresponding to the material's bandgap calculations. Below these thresholds, photon absorption is not observed, consistent with the material's electronic structure as described in [137]. Absorption increases progressively from ~ 3.5 to ~ 9.5 eV, with fluctuations and peaks that highlight interband transitions. These transitions, particularly in the ultraviolet region, play an essential role in *FeMnScAl*'s optical characteristics and suggest promising applications requiring strong ultraviolet absorption.

Narrow-bandgap materials with strong ultraviolet absorption are highly sought after in the field of optoelectronics, especially for devices such as LEDs, photodetectors, and solar cells, where efficient light absorption in thin films is essential [139,154].

The optical conductivity of *FeMnScAl* is examined in Figures A.24 (a) and (b), utilizing GGA and mBJ exchange-correlation potentials for both spin channels. For the majority spin state, illustrated in Figure A.34 (a), the $\sigma_1(\omega)$ values, beginning at zero eV, demonstrate the absence of an optical bandgap, highlighting metallic characteristics due to free carriers. This finding matches the behavior of the imaginary part of the dielectric function and the absorption coefficient for this spin state. Both GGA and mBJ methods reveal a notable rise in conductivity, with $\sigma_2(\omega)$ achieving peak values of $6.6 (\Omega \cdot m)^{-1}$

for GGA and $6.3 (\Omega\cdot m)^{-1}$ for mBJ at energy levels of 0.83 eV and 0.72 eV, respectively. After these peaks, the conductivity curves become oscillatory with several distinct peaks.

Figure A.24 (b) illustrates the imaginary part of the optical conductivity, $\sigma_2(\omega)$. Both curves exhibit fluctuations, alternating between negative and positive peaks. Such fluctuations of $\sigma_2(\omega)$ point out the dielectric nature of the material, and hence the peaks and oscillation are related to regions of polarizations affecting the way the light will propagate and, in general, store energy, as explained in [155].

In the spin-down state, as illustrated in Figure A.24 (c), the initial optical conductivity values, $\sigma_1(\omega)$, are 0.145 eV for GGA and 0.157 eV for mBJ. These thresholds correspond directly to the electronic bandgap energies determined from our band structure analysis, serving as independent validation of the calculated electronic properties. The consistency between electronic and optical findings highlights the reliability of our computational framework. The behavior of $\sigma_1(\omega)$ is closely linked to the imaginary part of the dielectric function, $\epsilon_2(\omega)$, as well as the absorption coefficient, $\alpha(\omega)$, within this spin channel. The GGA and mBJ methods both exhibit a notable enhancement in optical conductivity, with $\sigma_1(\omega)$ reaching maxima of $7.64 (\Omega\cdot m)^{-1}$ and $7.57 (\Omega\cdot m)^{-1}$ at photon energies of 8.8 eV and 8.4 eV, respectively. Beyond these peak values, the optical conductivity displays a sophisticated oscillatory behavior, distinguished by sharp resonances indicative of the electronic band structure and multiple interband transitions. High $\sigma_1(\omega)$ values in particular photon energy ranges underline the material's capacity for significant light absorption and efficient charge carrier generation, emphasizing its potential for use in photovoltaic and optoelectronic technologies [156].

As shown in Figure A.24 (d), the imaginary component of the optical conductivity, $\sigma_2(\omega)$, demonstrates an oscillatory trend, oscillating between positive and negative values. This oscillatory behavior points to dynamic variations in the material's dielectric properties over the photon energy range. The peaks in $\sigma_2(\omega)$ indicate areas of significant polarization, which are fundamentally important for assessing the refractive index and the material's energy storage capacity. Dispersion properties, strongly dependent on $\sigma_2(\omega)$, are of great relevance for understanding light propagation, as well as for

characterizing the material's potential for capacitive energy storage, because phase shifts are strongly correlated to the imaginary conductivity [155].

The reflectivity of *FeMnScAl*, calculated for both spin-up and spin-down states, is illustrated in Figures A.25 (a) and A.35 (b) using GGA and mBJ methods. In the spin-up state, the static reflectivity is 76% for GGA and 69% for mBJ, with both approaches reaching a maximum of 72% in the infrared region. After reaching this maximum, reflectivity decreases, oscillates, and eventually stabilizes between 35% and 50%. For the spin-down state, the static reflectivity values are 70% for GGA and 69% for mBJ, with peaks around 75% in the infrared region for both methods. Following these peaks, the reflectivity declines in a similar way, demonstrating oscillatory behavior, and stabilizes between 36% and 53% in the visible and ultraviolet parts of the spectrum.

Reflectivity, conductivity, and absorption exhibit an inverse relationship, emphasizing the interplay between these properties. The behaviors of spin-dependent reflectivity and absorption suggest potential use in advanced spintronic technologies, such as detectors for spin-polarized light and spin filters. Figures A.24 (b) and (d) reveal the oscillations of $\sigma_2(\omega)$, which emphasize the material's changing dielectric properties and its applicability to cutting-edge optical and spintronic devices [138,139,157].

The energy loss function, $L_{ij}(\omega)$, for the spin-up and spin-down channels is shown in Figures A.26 (a) and A.26 (b). In Figure A.26 (a), which corresponds to the spin-up channel, $L_{ij}(\omega)$ begins at zero for low photon energies and gradually increases as photon energy rises. Oscillations superimposed on this rising trend result from electromagnetic waves interacting with the material's electronic structure, with collective excitations like plasmons being the main contributors to its optical and electronic characteristics. Conversely, Figure A.26 (b), showing the spin-down channel, reveals no absorption below approximately 0.145 eV (GGA) and 0.157 eV (mBJ). The observation of negligible energy loss at lower photon energies implies that electromagnetic radiation has little impact on *FeMnScAl* in this range, probably because no electronic transitions are available. The gradual increase in energy loss observed in both curves at elevated photon energies suggests an enhanced interaction with the electromagnetic field, predominantly driven by plasmon resonances and electronic transitions. Additionally, small peaks and fluctuations in the curves correspond to distinct energy loss events and specific electronic transitions. The overall rise in energy loss at elevated photon

energies emphasizes the significance of collective electron dynamics and high-energy excitations in shaping the material's optical behavior. Table 8 provides a summary of FeMnScAl's critical optical properties for both spin-up and spin-down states, derived from GGA and mBJ calculations.

Table 9

Material characterization matrix: spin states and methods

Property method	Spin-Up ↑		Spin-Down ↓	
	GGA	mBJ	GGA	mBJ
Dielectric Properties				
Static Dielectric Constant $\epsilon_1(0)$	217.67	129.13	120.00	129.13
Peak $\epsilon_2(\omega)$ Energy	0.28 eV	0.31 eV	0.45 eV	0.37 eV
Refractive Properties				
Static Refractive Index $n(0)$	11.36	14.73	10.80	11.26
Peak $n(\omega)$ Energy	0.05 eV	0.23 eV	0.20 eV	0.28 eV
Conductivity				
Peak $\sigma_1(\omega)$ Value $(\Omega \cdot m)^{-1}$	6.60	6.30	7.64	7.57
Peak $\sigma_1(\omega)$ Energy	0.83 eV	0.72 eV	8.80 eV	8.40 eV
Optical Bandgap	0	0	0.145 eV	0.157 eV
Reflectivity				
Static Reflectivity	76%	69%	70%	69%
Maximum Reflectivity	72%	72%	75%	75%
Stabilized Range (LV-UV)	35-50%	35-50%	36-53%	36-53%

3.2.8 Thermoelectric Properties

Figure A.27 illustrates the material's thermoelectric characteristics, divided into spin-up and spin-down channels, within the energy range $E - E_F = 5.03 \times 10^{-3}$ eV. The spin-up electronic density of states ($N\uparrow$), shown in Figure A.27 (a), gradually decreases with rising temperature, a pattern consistently seen in both GGA and mBJ approaches. Like $N\uparrow$, the spin-down electronic density ($N\downarrow$), depicted in Figure A.27 (b), shows a decrease with temperature in both GGA and mBJ approaches. Figure A.27 (c) illustrates the spin-up Seebeck coefficient ($S\uparrow$), which similarly decreases with temperature according to both methods. The presence of *p*-type and *n*-type charge carriers within

this temperature range is indicated by the positive and negative values of $S\uparrow$, respectively.

Figure A.37 (d) represents how the Seebeck coefficient for the spin-down channel ($S\downarrow$) varies with temperature. The value of $S\downarrow$ initially declines as the temperature rises to 200 K, peaks at $196 \mu V K^{-1}$ at 300 K, and then shows a gradual upward trend as the temperature increases to 500 K. The positive $S\downarrow$ values indicate that p-type charge carriers are predominant in this temperature interval. As seen in Figure A.27 (e), the spin-up electrical conductivity ($\sigma\uparrow$) decreases at first and then begins to increase with temperature. The GGA and mBJ methods show close agreement at lower temperatures, but minor discrepancies emerge as temperature increases. In Figure A.27 (f), the spin-down electrical conductivity ($\sigma\downarrow$) rises consistently with temperature for both methods. The electronic thermal conductivity of the spin-up channel ($\kappa_e\uparrow$), shown in Figure A.27 (g), increases steadily as the temperature rises. Likewise, Figure A.27 (h) demonstrates that $\kappa_e\downarrow$ for the spin-down channel follows a similar upward trend in both GGA and mBJ calculations. The *FeMnScAl* alloy analysis reveals differences in electrical conductivity based on spin state, with the spin-up channel displaying higher conductivity near the Fermi level, which aligns with its half-metallic characteristics. For both spin states, electronic conductivity generally increases as temperature rises. The thermoelectric properties for the spin-up ($\uparrow$) and spin-down ($\downarrow$) channels at particular temperatures are detailed in Tables 10.

Table 10

Spin-up thermoelectric properties at varying temperatures

Property Method	50K (GGA)	50K (mBJ)	300K (GGA)	300K (mBJ)	500K (GGA)	500K (mBJ)
$N\uparrow$	-3.0149	-3.0154	-3.0185	-3.0206	-3.0304	-3.0356
$S\uparrow$ (eV/K)	2.19×10^{-4}	2.16×10^{-4}	-203.7	-252	-420	-465.15
$\sigma\uparrow$ (1/ ΩmsK)	1.08×10^2	1.09×10^2	8.1×10^1	8.16×10^1	8.76×10^1	8.91×10^1
$\kappa_e\uparrow$ (W/mK)	8.96×10^{13}	8.33×10^{13}	6.7×10^{13}	6.94×10^{13}	1.51×10^{14}	1.56×10^{14}
$N\downarrow$	2.9629	2.98436	2.85302	2.8897	2.73915	2.78327
$S\downarrow$ (eV/K)	-1.03×10^{-4}	1.6×10^{-4}	-1.8×10^{-4}	-2×10^{-4}	-1.51×10^{-4}	-1.6×10^{-4}
$\sigma\downarrow$ (1/ ΩmsK)	1.04×10^1	4.68×10^{18}	2.64×10^1	2.11×10^1	4.30×10^1	3.69×10^1
$\kappa_e\downarrow$ (W/mK)	1.63×10^{13}	1.04×10^{13}	4.38×10^{13}	3.86×10^{13}	1.25×10^{14}	1.15×10^{14}

Thermoelectric parameters, including the Figure of merit (ZT) and power factor (PF), are computed and shown in Figure A.28 (a-d). As shown in Figure A.28 (a), the spin-up $ZT_{\uparrow}$ declines with rising temperatures, hitting its lowest point at 100 K. Beyond this temperature, it begins to rise consistently and maintains its upward movement until 500 K. For the spin-down configuration, as depicted in Figure A.28 (b), the Figure of merit ($ZT_{\downarrow}$) initially rises with temperature, peaking at a value of 0.689 at 150 K under the mBJ approximation. However, above this maximum, $ZT_{\downarrow}$ gradually declines as the temperature continues to increase.

According to Figure A.28 (c), $PF_{\uparrow}$ for the spin-up state rises with temperature for both GGA and mBJ approximations, reaching its maximum near 400 K and then dropping slightly. On the other hand, Figure A.28 (d) shows that $PF_{\downarrow}$ for the spin-down configuration initially declines, hitting a minimum at 100 K, but later increases sharply with temperature, indicating better power generation potential at higher temperatures for both spins. At increased temperatures, both spin-up and spin-down PF values show trends that are highly consistent with ZT . Specifically, the spin-down ZT value for the *FeMnScAl* alloy at 300 K demonstrates promising potential for thermoelectric applications.

Notably, the *FeMnScAl* alloy's spin-down ZT value at 300K reveals promising properties for thermoelectric performance. A detailed presentation of ZT and PF values for different temperatures is included in Table B.11.

The Hall coefficient (R_H) of the *FeMnScAl* alloy, presented in Figure A.29, is consistently negative over the full temperature range in both spin-up and spin-down configurations, signifying n-type (electron-dominated) conduction. For the spin-up channel, R_H first declines to its lowest value at 100 K with mBJ and at 150 K with GGA, after which it rises as the temperature increases. For the spin-down channel, R_H exhibits a steeper upward trend, culminating in a peak at 500 K. The constant negative values of R_H throughout the temperature range for both spin configurations strongly indicate that electrons (n-type) are the main charge carriers [143]. Table B.1 includes the detailed R_H values.

The *FeMnScAl* alloy's lattice thermal conductivity (κ_l) was assessed through the Slack model, where transverse (V_t) and longitudinal (V_l) elastic wave velocities were integral

parameters. Using WIEN2k's IRelast module, the Debye temperature (θ_D) and Grüneisen parameter (γ) were computed [36,106]. The temperature-dependent variations in lattice and total thermal conductivity are depicted in Figures A.30 (a) and (b). The lattice thermal conductivity κ_l undergoes a sharp reduction up to 200 K, probably due to intensified phonon scattering, before decreasing at a slower rate until 500 K. At 300 K, it is recorded as 2.079 W/mK. The measured value at 300 K is 2.079 W/mK. Figure A.30 (b) illustrates that κ_{tot} for the spin-down state rises with temperature, following a pattern comparable to κ_e under both GGA and mBJ approximations. This trend suggests that the lattice component plays a minor role. The spin-down configuration was chosen because of its significance in electronic transition research.

This investigation reveals the significant utility of the *FeMnScAl* spin-down channel for thermoelectric purposes, particularly when its semiconducting nature is evident. This material's potential is clearly illustrated by the Seebeck coefficient of $196 \mu\text{V K}^{-1}$ at 300 K and the peak Figure of merit ($ZT_{\downarrow}$) of 0.689 at 150 K, which outperforms the spin-up channel. These results underscore the critical role of the spin-down channel in advancing efficient waste heat recovery technologies [5,158].

3.3 FeMnScGa Compound

3.3.1 Structural Properties and Thermodynamic Stability

To explore the structural stability of *FeMnScGa*, a quaternary Heusler compound, a detailed investigation was undertaken. Exhibiting a face-centered cubic crystal structure, it falls under the *LiMgPdSn*-type and Fm-3m (#216) space group. Optimizing the unit cell volume enabled the identification of the configuration most stable under normal conditions. Three configurations, named Type 1, Type 2, and Type 3, were analyzed; while all maintain cubic symmetry, they vary in the assignment of Wyckoff positions for *Fe*, *Mn*, *Sc*, and *Ga*. In Type 1, *Fe* occupies the *4a* position (0, 0, 0), *Mn* is placed at *4c* (0.25, 0.25, 0.25), *Sc* is assigned to *4b* (0.5, 0.5, 0.5), and *Ga* resides at *4d* (0.75, 0.75, 0.75). Within the Type 2 configuration, *Fe* is situated at *4a* (0, 0, 0), *Mn* occupies the *4b* position (0.5, 0.5, 0.5), *Sc* is found at *4c* (0.25, 0.25, 0.25), and *Ga* resides at *4d* (0.75, 0.75, 0.75). In contrast, in Type 3, *Fe* is found at *4c* (0.25, 0.25, 0.25), *Mn* resides at *4b* (0.5, 0.5, 0.5), *Sc* is at *4a* (0, 0, 0), and *Ga* is situated at *4d* (0.75, 0.75, 0.75). Calculations of energy were performed to identify the arrangement with the

greatest stability [115–117]. Figure A.31 illustrates the three crystallographic structures of the *FeMnScGa* compound, accompanied by its Brillouin zone, reciprocal vectors, and high-symmetry points. Ground-state properties are calculated using DFT at 0 K, effectively removing the influence of Fermi-Dirac occupation probabilities, with a zero Pascal vacuum to avoid external effects [159]. A volume optimization method was employed to systematically investigate these structural arrangements. In accordance with the Murnaghan equation of state, this approach represents total energy and pressure as dependent on volume per formula unit, as outlined in Equation (57). The energy-volume relationship obtained from geometry optimization, as illustrated in Figure A.32, confirms that the Type 2 arrangement with Fe at 4a, Mn at 4b, Sc at 4c, and Ga at 4d is the most stable configuration of *FeMnScGa*. As presented in Table B.4, our DFT calculations yield an optimized lattice parameter of 6.106 Å and a bulk modulus of 118.28 GPa, which are in close agreement with the reported values of 6.02 Å, 6.10 Å, and 120.41 GPa [144,160].

Equation (59) defines the formation energy E_{Form} of *FeMnScGa* by calculating the difference between the compound's total ground state energy and the sum of the ground state energies of *Fe*, *Mn*, *Sc*, and *Ga* in their standard forms [29].

$$E_{Form} = E_{FeMnScGa(x)}^{Cubic} - (E_{Fe}^{Cubic} + E_{Mn}^{Cubic} + E_{Sc}^{Cubic} + E_{Ga}^{Cubic}) \dots\dots\dots (60)$$

The energy $E_{FeMnScGa(x)}^{Cubic}$ represents the compound's total energy, whereas E_{Fe}^{Cubic} , E_{Mn}^{Cubic} , E_{Sc}^{Cubic} , and E_{Ga}^{Cubic} denote the energies of *Fe*, *Mn*, *Sc*, and *Ga*, respectively. The subscript (x = 1, 2, or 3) distinguishes among the three configurations. The negative formation energy in Table B.3 verifies the thermodynamic stability of *FeMnScGa*.

By calculating the cohesive energy, the structural stability of *FeMnScGa* across its three configurations is evaluated. This is done by normalizing the energy difference between the compound's total energy and the energies of its isolated constituent atoms, divided by the number of atoms in the unit cell.

$$E_{coh.} = \frac{(E_{Fe}^{iso} + E_{Mn}^{iso} + E_{Sc}^{iso} + E_{Ga}^{iso}) - E_{FeMnScGa(x)}^{Cubic}}{4} \dots\dots\dots (61)$$

Serving as an important stability indicator, cohesive energy reflects energetically favorable configurations with positive values. As detailed in Table B.3, these results,

alongside mechanical stability evaluations, reinforce the compound's strong potential for progressing research and technological development.

Only the *FeMnScGa* Type 2 structure will be analyzed for elastic, electronic, and optical properties because it is both thermodynamically and mechanically stable and represents the configuration with the lowest energy.

3.3.2 Phonon Analysis of FeMnScGa's Dynamical Stability

To evaluate the dynamic stability of the investigated compound, Quantum ESPRESSO's linear response method was employed to compute its phonon dispersion [161], and the corresponding phonon dispersion curve is illustrated in Figure A.33. A compound is considered dynamically stable if its phonon spectrum lacks soft modes (or imaginary frequencies, represented as negative values). Dynamic instability is reflected by the occurrence of soft modes, which may result in structural phase transitions and lattice instabilities. Additionally, this could suggest that synthesizing the material under typical conditions is challenging. As illustrated in Figure A.33, the phonon spectrum of FeMnScGa does not contain soft modes, thereby validating its dynamic stability [122,146–149]. As the structure with the lowest energy and confirmed thermodynamic and dynamic stability, FeMnScGa type 2 is the best-suited candidate for investigating its elastic, electronic, magnetic, optical, and thermoelectric properties.

3.3.3 Elastic Properties

Mechanical stability, in conventional terms, requires that a material's elastic constants fulfill the conditions $C_{11} > 0$, $C_{11} + 2C_{12} > 0$, $C_{12} < B < C_{11}$, $C_{11} - C_{12} > 0$, and $C_{44} > 0$. *FeMnScGa* satisfies these criteria, confirming its mechanical stability, as shown in Table B.4 (elastic constants at zero strain) [119]. The characteristics of stiffness and shear resistance were quantified by employing Young's modulus and shear modulus [37]. A Poisson's ratio of 0.317, surpassing the threshold of 0.26 for ductility, reflects the material's ductile nature [38]. Poisson's ratio also differentiates bond types, with values under 0.25 implying ionic bonds and values over 0.25 indicating covalent bonds. The ratio calculated in the study validates covalent bonding [44]. Mechanical behavior variations, influenced by loading direction, are captured by the anisotropic factor, with $A = 1$ indicating isotropy and $A > 1$ denoting anisotropy [37,47,48]. Using elastic constants and average sound velocity ($v_{\square}$), the Debye temperature (Θ_D) is calculated

[48,121]. A high Debye temperature and melting point range of 1266.0254–1866.0254 K suggest that *FeMnScGa* type 2 exhibits remarkable thermal stability and can endure high temperatures, making it suitable for high-temperature and mechanically demanding applications [48]. *FeMnScGa* type 2 exhibits properties that indicate strong performance under mechanical stress and high-temperature conditions.

3.3.4 Electronic Properties

The electronic properties of elements, as well as their alloys, are essential for comprehending chemical bonding. This research focuses on the density of states (DOS) and band structure of the *FeMnScGa* compound type 2. Using GGA and mBJ approximations, Figures A.44 (a-e) and A.44 (a-e) depict the total and partial DOS, providing insight into the origin of the energy band gap. The d-orbitals of *Fe*, *Cr*, and *Sc* are the primary contributors to the DOS near the Fermi level in both spin states, whereas *p*- and *s*-orbitals have minimal influence in either spin channel. This compound exhibits half-metallicity with 100% spin polarization, making it suitable for spintronic applications.

Figure A.35 provides a representation of the spin-polarized electronic band structures of the *FeMnScGa* alloy, calculated using GGA and mBJ potentials. The spin-up and spin-down states, determined by GGA, are illustrated in Figures A.36 (a) and A.36 (b). Figures A.36 (a) and A.36 (b) illustrate the spin-up and spin-down states, as determined by GGA. Metallic behavior is observed in the spin-up state as bands cross E_F , while the spin-down state shows semiconducting properties with a band gap around E_F . Figures A.36 (c) and A.36 (d) display the more detailed electronic structure derived from the mBJ potential. The mBJ-up state, as is expected from the GGA-up result, still has metallic behavior since bands cross E_F , while the mBJ-down state shows more of an enhanced band gap. Both GGA and mBJ predict an indirect bandgap in the first Brillouin zone with GGA giving 0.189 eV and mBJ giving 0.442 eV with the valence band maximum at Γ and conduction band minimum at X. The combination of metallic and semiconducting behavior, a hallmark of half-metals, is essential for spintronic applications due to its ability to produce highly spin-polarized currents. The mBJ potential enhances the half-metallic nature of *FeMnScGa* by more accurately predicting the spin-down band gap.

3.3.5 Electronic Charge Density and Chemical Bonding

As a key method for visualizing chemical bonding, charge density reflects the interaction range between atoms based on electron cloud distribution and is widely used in charge distribution studies. The (110) plane of FeMnScGa is represented in Figure A.37 (a-b), showing its 2D charge density contours for both majority and minority spin states. Around Fe and Mn atoms, spherical charge distributions indicate the ionic nature of the bonds [123,124]. In contrast, the covalent nature of bonds is apparent in the distorted and stretched contours around Sc and Ga atoms [125]. These distinctions are attributable to the differences in electronegativity: Fe (1.83), Mn (1.55), Sc (1.36), and Ga (1.81).

3.3.6 Magnetic Properties

Using GGA and mBJ methods, the magnetic characteristics of the *FeMnScGa* alloy were determined, as shown in Table B.4. The total magnetic moment of about 3 μB confirms its half-metallic behavior and complete spin polarization. The major contribution to the total magnetic moment originates from the Mn atom, whose high magnetic moment is attributed to its half-filled $3d^5$ configuration. This configuration favors a high-spin state according to Hund's rule, thereby promoting ferromagnetic alignment, while Fe adds a smaller positive moment, and Sc and Ga contribute negatively. The interstitial region also exhibits a minor negative moment in both methods. The total spin magnetic moment aligns with the Slater-Pauling rule, which connects the total magnetic moment (M_{tot}) to the total valence electron count (Z_{tot}) via $M_{\text{tot}} = Z_{\text{tot}} - 24$. The valence electron contributions for *FeMnScGa* are Fe: 8 ($3d^6 4s^2$), Mn: 7 ($3d^5 4s^2$), Sc: 3 ($3d^1 4s^2$), and Ga: 3 ($4s^2 4p^1$), resulting in 21 valence electrons and a magnetic moment of 3 μB . This feature emphasizes its potential for spintronics and other high spin-polarization applications.

The stability and applications of magnetic materials rely on a detailed understanding of their properties in different ground states. By examining total energies and structural parameters, FM, PM, and AFM states can be accurately evaluated within the GGA-PBE framework. This assessment is achieved through the analysis of total energies and structural parameters. The enhanced stability of the FM state, attributed to its lower energy and favorable structural features, is confirmed by data from Table B.5 and Figure A.37. Through a detailed representation of energy variations, Figure A.37

provides strong evidence supporting this conclusion. Superior structural adaptability, indicated by a reduced bulk modulus, makes the FM state the most favorable configuration.

Magnetic material stability and applications require in-depth analysis of the distinct properties associated with various ground states. The GGA-PBE framework offers a reliable approach for assessing ferromagnetic (FM), paramagnetic (PM), and antiferromagnetic (AFM) states through comparisons of total energy and structural parameters. The lower energy and superior structural characteristics of the FM state, as demonstrated in Table B.6 and Figure A.38, contribute to its enhanced stability. Include this final statement only if the disparities highlighted in Figure A.38 are the primary focus, rather than merely serving as a visual representation of the lower energy already discussed. GGA-PBE calculations show the FM state has a lower bulk modulus. This indicates superior structural adaptability, making the FM state the optimal configuration.

3.3.7 Optical properties

The spectroscopic investigation of *FeMnScGa*'s dielectric function, ranging from infrared to ultraviolet energies (0-8 eV), is documented in Figure A.39. Superior electric field neutralization capabilities are demonstrated by the spin-up state's elevated static dielectric constants, calculated as 196.21 (GGA) and 154.32 (mBJ). The static dielectric constant measures how effectively a material can counteract electric fields produced by trapped charges, making it a crucial parameter for assessing its response to low-frequency or static electric fields [152]. A shift from metallic to dielectric behavior is marked by the first plasma frequency (ω_p), where $\epsilon_1(\omega_p) = 0$, observed at 0.286 eV (GGA) and 0.422 eV (mBJ). Infrared peaks in $\epsilon_1(\omega)$ suggest strong optical performance, while negative $\epsilon_1(\omega)$ values in parts of the infrared and ultraviolet spectra highlight metallic characteristics, emphasizing *FeMnScGa*'s suitability for optical filtering and electromagnetic shielding. The imaginary component of the dielectric function, $\epsilon_2(\omega)$ (Figure A.39 (b)), characterizes absorption. For both GGA and mBJ, the peaks in $\epsilon_2(\omega)$ are located at 215 and 214, corresponding to energy values of 0.17 eV and 0.34 eV, respectively. The $\epsilon_2(\omega)$ values, which start at zero eV, indicate the absence of an optical band gap, confirming the material's metallic nature.

In Figure A.39 (c), the spin-down state's $\epsilon_1(\omega)$ curve displays a static dielectric constant ($\epsilon_1(0)$) of 108.42 (GGA) and 75.79 (mBJ). Optical transitions are represented by peaks at 0.32 eV (GGA) and 0.55 eV (mBJ). These peaks, which decline sharply in the infrared region, result from inter-band transitions of bonding electrons, indicating strong interactions between energy bands in this spectrum. Figure A.39 (d) presents the imaginary component of the dielectric function, denoted as $\epsilon_2(\omega)$, which represents the material's absorption of radiation. The $\epsilon_2(\omega)$ peaks are located at 0.39 eV (GGA) and 0.64 eV (mBJ). $\epsilon_2(\omega)$ remains at zero until the photon energy exceeds the band gap, after which absorption initiates. This defines the energy threshold for a direct optical transition, facilitating electron excitation from the valence band maximum to the conduction band minimum.

The refractive index $n(\omega)$ and extinction coefficient $k(\omega)$ for FeMnScGa are depicted in Figure A.39, with data separated into majority and minority spin channels. Results for the majority spin are shown in Figures A.40 (a) and (b), whereas Figures A.40 (c) and (d) present calculations for the minority spin using the GGA and mBJ methods. Figures A.40 (a) and (b) display the computed $n(\omega)$ and $k(\omega)$ values for the majority spin channel using GGA and mBJ approaches. Static refractive indices, 14.01 for GGA and 12.42 for mBJ, satisfy the equation $n(0) = \sqrt{\epsilon_1(0)}$, which relates $n(\omega)$ to the real part of the dielectric function $\epsilon_1(\omega)$ [132]. The maximum refractive index for GGA occurs at 15.08 at 0.15 electron volts, whereas for mBJ, it peaks at 16.644 at 0.204 electron volts, with both declining as photon energy increases. In the ultraviolet spectrum, *FeMnCrGe* has the lowest $n(\omega)$. The extinction coefficient $k(\omega)$, illustrated in Figure A.40 (b), is directly influenced by the imaginary part of the dielectric function.

Figures A.40 (c) and A.50 (d) show how the refractive index, $n(\omega)$, and the extinction coefficient, $k(\omega)$, change with photon energy in the minority spin channel. Figure A.40 (c) gives static $n(\omega)$ values of 7.72 for GGA and 9.46 for mBJ, matching the theoretical relationship between $n(0)$ and $\epsilon_1(\omega)$ [132]. Peaks in $n(\omega)$ appear in the infrared region at about 0.45 eV for GGA and 0.53 eV for mBJ, paralleling the behavior of $\epsilon_1(\omega)$. In the visible range, $n(\omega)$ stays elevated, varying from 2.56 to 4.18, before decreasing gradually as photon energy increases, with comparable results from both methods. Figure A.39 (d) shows that $k(\omega)$ closely follows $\epsilon_2(\omega)$, staying at zero until the photon energy exceeds the band gap, which starts absorption. The sharp peaks in $k(\omega)$ match

strong optical absorption and transitions caused by photon energy. A high extinction coefficient is seen at lower energy levels, with clear peaks around 1.67 eV (GGA) and 1.04 eV (mBJ), indicating significant absorption events.

As shown in Figures A.41 (a) and A51 (b), the optical absorption of *FeMnScGa* is analyzed using the GGA and mBJ exchange-correlation potentials for the majority and minority spin channels. In the spin-up channel, as shown in Figure A.40 (a), absorption exhibits metallic characteristics, initiating at low photon energies and progressively increasing with higher photon energy. Figures A.41 (a) and A.41 (b) illustrate the optical absorption of *FeMnScGa*, which has been analyzed using the GGA and mBJ exchange-correlation potentials for the majority and minority spin channels. As expected, photon absorption is absent below these energy thresholds, reflecting the underlying electronic structure [137]. The absorption intensifies with rising photon energy, displaying fluctuations and peaks that mark transition regions dominated by interband transitions. Particularly significant in the ultraviolet range, these transitions emphasize *FeMnScGa*'s optical capabilities and potential applications in ultraviolet absorption. Narrow-bandgap materials with robust UV absorption are highly valued for optoelectronic devices like LEDs, photodetectors, and solar cells, where effective light absorption in thin films is crucial [139,154].

The optical conductivity of *FeMnScGa* is illustrated in Figures A.42 (a) and A.42 (b), with calculations performed using GGA and mBJ exchange-correlation potentials for majority and minority spin channels. As shown in Figure A.42 (a), $\sigma_1(\omega)$ in the majority spin state begins at zero eV, confirming the absence of an optical bandgap and indicating metallic properties caused by free carriers. This behavior aligns with the imaginary dielectric function and absorption coefficient in this spin state. Both GGA and mBJ exhibit a steep rise in the curves, with $\sigma_2(\omega)$ peaking at $5.21 (\Omega.m)^{-1}$ and $8.92 (\Omega.m)^{-1}$ at 0.23 eV and 0.37 eV, respectively, followed by oscillations with prominent peaks. The optical conductivity's imaginary component $\sigma_2(\omega)$, depicted in Figure A.42 (b), exhibits alternating maxima and minima in its oscillatory pattern. These observed fluctuations in $\sigma_2(\omega)$ demonstrate consistency with the dielectric characteristics, revealing domains of polarization that influence both photonic propagation and energy storage [155].

For the spin-down state, as shown in Figure A.42(c), the $\sigma_1(\omega)$ values begin at 0.189 eV (GGA) and 0.442 eV (mBJ), corresponding precisely to the optical conductivity thresholds. These values are in agreement with the electronic band gap energies derived from our band structure calculations for this spin channel. This serves as independent validation of our computed electronic structure. The alignment between the electronic and optical properties highlights the accuracy of our computational model. The behavior of $\sigma_1(\omega)$ is closely linked to both $\epsilon_2(\omega)$, the imaginary part of the dielectric function, and the absorption coefficient, in this spin state. The GGA and mBJ calculations indicate a sharp rise in optical response, with the real conductivity, $\sigma_1(\omega)$, showing significant peaks at $10.07 (\Omega\cdot m)^{-1}$ and $14.86 (\Omega\cdot m)^{-1}$, corresponding to photon energies of 0.612 eV and 1.02 eV. After these initial peaks, the optical conductivity reveals a complex oscillatory behavior, characterized by sharp resonances that highlight the influence of the material's electronic band structure on interband transitions. The high $\sigma_1(\omega)$ values in specific energy ranges indicate strong light absorption and effective free charge carrier generation, which enhances the material's prospects for use in photovoltaic and optoelectronic devices [156]. Figure A.42 (d) demonstrates the oscillatory nature of $\sigma_2(\omega)$, the imaginary part of the optical conductivity, which fluctuates between positive and negative values, signaling changes in the dielectric properties across the photon energy range. Peaks in $\sigma_2(\omega)$ indicate regions of strong polarization, affecting both the refractive index and the material's energy storage capacity [155]. Understanding the dispersion properties of $\sigma_2(\omega)$ is critical for evaluating light propagation in the material and its applicability in energy storage for capacitive devices, with the imaginary conductivity playing a key role in determining phase shifts.

Figures A.43 (a) and (b) show the reflectivity of *FeMnScGa* for spin-up and spin-down states, computed using the GGA and mBJ methods. For the spin-up state, the static reflectivity values are 75% (GGA) and 72% (mBJ), peaking at 80% in the infrared region. Beyond the peak, the reflectivity decreases, shows oscillatory behavior, and eventually stabilizes between 40% and 55%. For the spin-down state, the static reflectivity is 59% (GGA) and 65% (mBJ), with peaks reaching around 71% (GGA) and 80% (mBJ) in the infrared region. Similar to the spin-up state, the reflectivity for the spin-down state also decreases, fluctuates, and stabilizes between 40% and 55% in the visible and ultraviolet regions. A distinct inverse relationship is observed between

reflectivity and both conductivity and absorption. The spin-dependent reflectivity and absorption indicate potential applications in spintronic devices, such as spin filters and spin-polarized light detectors. Changes in the material's dielectric properties across the photon energy range are reflected in the oscillatory behavior of $\sigma_2(\omega)$, the imaginary part of optical conductivity, which alternates between positive and negative values, as shown in Figures A.38 (b) and A.38 (c) [138,139,157].

The energy loss function, $L(\omega)$, for both spin channels is presented in Figures A.54 (a) and A.54 (b). For the spin-up state, as shown in Figure A.54 (a), $L(\omega)$ begins at zero for low photon energies and progressively rises as the photon energy increases. This gradual increase is accompanied by oscillations resulting from the interaction of electromagnetic waves with the material's electronic structure. These oscillatory patterns are mainly associated with collective excitations, such as plasmons, which are critical in shaping the material's optical and electronic properties.

As illustrated in Figure A.44 (b), the spin-down state exhibits no energy loss below roughly 0.189 eV for GGA and 0.442 eV for mBJ. This negligible loss implies that *FeMnCrGe* is generally unaffected by electromagnetic radiation at these energies, likely due to the unavailability of electronic transitions. As photon energies rise, energy loss in both methods becomes more pronounced, reflecting enhanced interactions between the material and the electromagnetic field. Such dissipation results from phenomena like electronic transitions and plasmon resonances.

At certain photon energies, discrete energy loss events and distinct electronic transitions appear as small peaks and irregularities in the energy loss curves. The consistent increase in energy loss as photon energy grows emphasizes the influence of collective electronic behavior and high-energy phenomena on the material's interaction with electromagnetic radiation.

The results demonstrate that FeMnScGa, with its narrow bandgap, high refractive index, low reflectivity across a wide range from visible to ultraviolet, high optical conductivity, and strong ultraviolet absorption, is an excellent material for enhanced light trapping in photovoltaic applications. A comprehensive analysis of key optical properties, along with their respective energy values, has been conducted using GGA and mBJ calculations, with the results compiled in Table B.7

Optimal characteristics for photovoltaic light-trapping applications are exhibited by FeMnScGa, as confirmed by comprehensive analysis. The material's properties include a narrow bandgap, a high refractive index, low reflectivity across a broad range from visible to ultraviolet light, excellent optical conductivity, and significant ultraviolet absorption. Table B.6 offers a detailed summary of these features, calculated using both GGA and mBJ methods, along with their corresponding energy values.

3.3.8 Thermoelectric Properties

The thermoelectric properties of the FeMnScGa alloy are investigated in this section, with a focus on the spin-up and spin-down channels. Two approximation methods, GGA and mBJ, are utilized to analyze these properties near the Fermi level ($E - E_F = 18 \times 10^{-4}$ eV). The thermoelectric properties of spin-up and spin-down channels, within the 50 to 600 K temperature range, are evaluated by the study using GGA and mBJ methods, as depicted in Figure A.45 (a-h). Figure A.45 (a) emphasizes the relationship between temperature and spin-up carrier concentration ($N_{\uparrow}$), providing essential insights into the electronic and thermal properties of the material. The carrier concentration $N_{\uparrow}$ decreases with increasing temperature for both approximations. At 50 K, $N_{\uparrow}$ is -0.52659 for GGA and -0.54317 for mBJ. By 300 K, these values drop to -0.50650 (GGA) and -0.52267 (mBJ), and further to -0.48847 (GGA) and -0.50286 (mBJ) at 600 K, indicating thermal excitation's role in reducing carrier density near the Fermi level. Figure A.45 (b) shows a similar trend for the spin-down carrier concentration ($N_{\downarrow}$), which decreases from 0.54828 (GGA) and 0.54677 (mBJ) at 50 K to 0.54089 (GGA) and 0.53962 (mBJ) at 300 K, and finally to 0.51510 (GGA) and 0.51382 (mBJ) at 600 K.

Figure A.45 (c) illustrates the spin-up Seebeck coefficient ($S_{\uparrow}$), which increases sharply at lower temperatures before plateauing at higher temperatures. The Seebeck coefficient values calculated using GGA and mBJ display a consistent upward trend at 200 K, 300 K, and 600 K. At 200 K, GGA yields 24.5 $\mu\text{V/K}$ for $S_{\uparrow}$, whereas mBJ gives a slightly higher value of 24.9 $\mu\text{V/K}$. These values increase to 31.9 $\mu\text{V/K}$ and 32.4 $\mu\text{V/K}$ at 300 K for GGA and mBJ, respectively, reaching 39.8 $\mu\text{V/K}$ and 40 $\mu\text{V/K}$ at 600 K. The positive $S_{\uparrow}$ values suggest that p-type charge carriers, particularly holes, dominate charge transport. According to Figure A.45 (d), the spin-down Seebeck coefficient ($S_{\downarrow}$) is relatively small at 50 K, not exceeding 9.58 $\mu\text{V/K}$ for either method, which aligns

with the tendency for lower values at reduced temperatures. At 300 K, $S_{\downarrow}$ rises to 12.76 $\mu\text{V/K}$ for GGA and 12.82 $\mu\text{V/K}$ for mBJ, indicating enhanced thermoelectric performance and strong agreement between the methods. At 600 K, the coefficient peaks at around 20.51 $\mu\text{V/K}$ for GGA and 20.5486 $\mu\text{V/K}$ for mBJ, showing minimal variation between the two approaches. The positive $S_{\downarrow}$ values indicate that holes (*p*-type) dominate as charge carriers.

The spin-up channel's temperature-dependent electrical conductivity ($\sigma/\tau_{\uparrow}$) can be observed in Figure A.45 (e). Conductivity peaks at 50 K, with the GGA method resulting in $3.1 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$, whereas the mBJ approximation yields a lower value of $2.9 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$, reflecting enhanced carrier scattering in the mBJ approach. At 300 K, the conductivity data illustrate the effect of temperature on carrier mobility, with GGA calculations decreasing to $2.87 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$ and mBJ predictions giving $2.82 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$. When the temperature increases to 600 K, thermal excitation is observed to play a role, causing a small increase in conductivity, with GGA and mBJ values rising to $2.88 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$ and $2.86 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$, respectively. These variations highlight the significant effect of the chosen approximation method on the material's thermoelectric properties, particularly its influence on conductivity calculations.

Figure A.45 (f) presents the temperature dependence of the spin-down channel's electrical conductivity ($\sigma/\tau_{\downarrow}$). At 50 K, GGA calculates a conductivity of $4.06898 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$, and mBJ predicts $4.04706 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$. This small variation implies that mBJ leads to a slightly higher level of carrier scattering, yet the conductivity remains significantly high. Thermal effects become apparent at 300 K, where increased carrier mobility leads to conductivity values of $4.05906 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$ for GGA and $4.05593 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$ for mBJ, with their close agreement indicating reduced sensitivity to approximation method selection. Further heating to 600 K results in decreased conductivity— $4.01737 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$ for GGA and $4.01644 \times 10^2 \square \text{ m}\cdot\text{s}/(\Omega\cdot\text{eV})$ for mBJ as thermal energy enhances carrier scattering beyond the benefits of additional carrier generation. The convergence of results at this temperature reinforces that thermal effects, not approximation method choice, dominate the spin-down channel's conductivity behavior.

Figure A.45 (g) shows the temperature-dependent electronic thermal conductivity ($\kappa_{\uparrow}$) for the spin-up channel, with GGA and mBJ calculations. The results reveal a temperature-induced increase in conductivity, highlighting the significance of thermal excitation in electronic transport. The methods yield distinctly different outcomes at 50 K, with GGA showing $1.48 \times 10^1 \text{ W/(m}\cdot\text{K)}$ and mBJ predicting $3.47 \times 10^1 \text{ W/(m}\cdot\text{K)}$, indicating that mBJ overestimates thermal conductivity at low temperatures because of its treatment of electronic interactions. Further temperature elevation to 600 K results in conductivity values of $3.67 \times 10^1 \text{ W/(m}\cdot\text{K)}$ and $4.51 \times 10^1 \text{ W/(m}\cdot\text{K)}$ for GGA and mBJ respectively, demonstrating thermal excitation's growing influence on electronic transport.

The temperature dependence of spin-down electronic thermal conductivity ($\kappa_{\downarrow}$) in the *FeMnScGa* alloy appears in Figure A.55 (h). Initial measurements at 50 K show thermal conductivity values of $5.42 \times 10^1 \text{ W/(m}\cdot\text{K)}$ using GGA and $1.06 \times 10^1 \text{ W/(m}\cdot\text{K)}$ using mBJ. Temperature elevation to 300 K produces linear conductivity increases, reaching $3.04 \times 10^1 \text{ W/(m}\cdot\text{K)}$ with GGA and $3.50 \times 10^1 \text{ W/(m}\cdot\text{K)}$ with mBJ. At 600 K, GGA predictions exceed mBJ estimates, showing values of $5.51 \times 10^1 \text{ W/(m}\cdot\text{K)}$ and $4.51 \times 10^1 \text{ W/(m}\cdot\text{K)}$ respectively, suggesting competing influences between thermal excitation and scattering mechanisms modeled differently by each approximation method. Both spin states demonstrate progressive enhancement of electronic conductivity with increasing temperature.

The spin-up channel's thermoelectric Figure of merit ($ZT_{\uparrow}$) analysis appears in Figure A.46 (a). Initial measurements at 50 K reveal $ZT_{\uparrow}$ values of 0.00417 and 0.00319 for GGA and mBJ, respectively. Temperature elevation to 300 K produces substantial efficiency improvements, with GGA yielding 0.04213 and mBJ showing a slightly higher value of 0.04292, demonstrating thermal effects' enhancement of thermoelectric performance. At 600 K, the methods converge to similar values: 0.06076 for GGA and 0.06094 for mBJ, indicating peak thermoelectric efficiency and reduced sensitivity to approximation method selection at elevated temperatures.

Figure A.46 (b) presents the spin-down channel's thermoelectric Figure of merit ($ZT_{\downarrow}$). The methods produce values of 0.00239 (GGA) and 0.00301 (mBJ) at 50 K, increasing to 0.00666 and 0.00672, respectively, at 300 K. Further temperature elevation to 600 K yields values of 0.01868 (GGA) and 0.01874 (mBJ), demonstrating thermal effects' growing influence on thermoelectric performance.

consistent enhancement of thermoelectric efficiency across both approximation methods.

The power factor ($PF\uparrow$) comparison for the spin-up state appears in Figure A.56 (c). At 50 K, GGA predicts 2.92×10^{11} W/mK² compared to mBJ's 2.21×10^{11} W/mK². Values increase substantially at 300 K to 2.91×10^{11} and 2.96×10^{11} W/mK² for GGA and mBJ, respectively. The methods yield nearly identical results at 600 K: 4.56×10^{11} W/mK² (GGA) and 4.58×10^{11} W/mK² (mBJ).

Temperature dependence of the spin-down power factor ($PF\downarrow$) is shown in Figure A.46 (d). Initial measurements at 50 K yield 2.29×10^{11} W/mK² using GGA and 2.88×10^{11} W/mK² using mBJ. Values grow to 6.61×10^{10} and 6.67×10^{10} W/mK² at 300 K, respectively, and continue to increase to 1.69×10^{11} and 1.70×10^{11} W/mK² at 600 K, showing consistent thermal effects across both methods.

In Figure A.47 (a), the spin-up Hall coefficient ($R_H\uparrow$) of the FeMnScAl alloy is compared using GGA and mBJ frameworks. Low-temperature measurements at 50 K show method-dependent variations, with mBJ yielding 7.05×10^{-11} m³/C and GGA producing a lower value of 6.40×10^{-11} m³/C. As temperature rises to 300 K, both methods show coefficient reduction, reaching 2.46×10^{-11} m³/C (GGA) and 2.37×10^{-11} m³/C (mBJ). The coefficients turn negative at 600 K, with GGA and mBJ values of -1.15×10^{-11} m³/C and -1.27×10^{-11} m³/C, respectively.

Figure A.47 (b) illustrates the spin-down Hall coefficient ($R_H\downarrow$), which shows a steady decline and increased negativity as temperature rises. At 50 K, GGA calculates -5.99×10^{-12} m³/C, whereas mBJ predicts -2.52×10^{-12} m³/C. At 600 K, the methods converge, with GGA and mBJ yielding nearly identical values of -2.14×10^{-11} m³/C and -2.13×10^{-11} m³/C. The *FeMnScGa* alloy's lattice thermal conductivity (κ_l) exhibits temperature-dependent behavior as calculated using the Slack model [100,101]. Figure A.48 reveals a non-linear temperature-dependent decline in κ_l , beginning at 1.93 at 50 K and diminishing to 0.161 at 600 K, with reduced decay rates at higher temperatures. Analysis of total thermal conductivity (κ_{Tot}) in Figures A.48 (a) and (b) reveals temperature-driven increases that closely follow the trends observed in electronic thermal conductivity (κ_e) for both GGA and mBJ frameworks. This similarity suggests that the contribution of lattice thermal conductivity is minimal.

Recognized for its rising Seebeck coefficient, strong power factor, and notable thermoelectric Figure of merit (ZT), the alloy exhibits great promise for thermoelectric applications. *FeMnScGa* is well-suited for energy harvesting and thermal management, especially in high-temperature environments where efficient thermal energy conversion is crucial. Table B.7 provides comprehensive documentation of these superior thermoelectric properties.

Chapter Four

Conclusions and Future Perspectives

This thesis presents a systematic study, based on accurate first-principles calculations, of three promising quaternary Heusler alloys: FeMnCrGe, FeMnScAl, and FeMnScGa. Accurate computational methods and a systematic study reveal detailed new information on the structural, electronic, magnetic, optical, and thermoelectric properties of these materials. With far-reaching implications for spintronics, energy conversion, and next-generation electronics, these findings radically transform our knowledge of these materials and their uses.

4.1 Structural Properties and Stability Analysis

These three compounds share a face-centered cubic structure (Fm-3m space group); however, their distinct stability profiles are critical in determining their potential for various applications. The Type 1 FeMnCrGe structure exhibited exceptional structural integrity, its negative formation energy indicating inherent thermodynamic stability. A lack of imaginary phonon modes throughout the Brillouin zone confirmed dynamic stability, and the notably high bulk modulus points to excellent compression resistance, essential for practical device use. The material's elastic constants met all mechanical stability criteria and, furthermore, demonstrated anisotropic behavior, which could be advantageous in directionally dependent applications.

The investigation of FeMnScAl revealed that its Type 2 configuration represents the optimal atomic arrangement, demonstrating both thermodynamic and mechanical stability. Charge density analysis reveals intriguing bonding characteristics in the compound. Spherical charge distributions around Fe and Mn indicate ionic bonds. Meanwhile, elongated contours around Sc and Al suggest substantial covalent interaction. The material's unique dual nature grants it outstanding mechanical flexibility and exceptional structural resilience, making it ideal for applications involving high stress.

The Poisson's ratio of 0.317 observed in the Type 2 FeMnScGa structure indicates its ductile nature, which could enhance fabrication processes. Optimal stability was attained by the material under zero temperature and pressure conditions. In addition, the

high Debye temperature reflected superior thermal stability, making it suitable for high-temperature applications. The compound's mixed ionic-covalent bonding characteristics, revealed through detailed charge density analysis, contribute to its balanced mechanical properties and structural resilience.

4.2 Electronic Structure and Magnetic Properties

FeMnCrGe exhibited remarkable spin polarization characteristics, achieving 100% spin polarization with near-perfect half-metallic behavior. The examination of electronic and magnetic properties detected sophisticated traits that advance these materials for spintronic applications. The material's electronic structure showed a distinct dichotomy between its spin channels, with the spin-up channel displaying metallic characteristics while the spin-down channel maintained a substantial semiconductor indirect band gap of 0.974 eV when calculated using the more accurate modified Becke-Johnson (mBJ) potential. This significant band gap suggests robust spin-polarized transport properties even at elevated temperatures. The magnetic properties of FeMnCrGe proved to be equally impressive, completely in line with Slater Pauling rules, with the total magnetic torque to 1.00 μ_B per formula unit. The strong magnetic interactions between Mn and Cr atoms underpin the material's robust ferromagnetic behavior, while the observed magnetic anisotropy suggests potential applications in magnetic storage devices. The temperature-dependent magnetic properties demonstrate stability well above room temperature. This stability is essential for practical applications.

FeMnScAl demonstrated exceptional half-metallic characteristics, with its spin-down channel featuring a bandgap of 0.1574 eV (mBJ) and a total magnetic moment of 3.00 μ_B per formula unit. Mn atoms play a dominant role in magnetic interactions, as indicated by a thorough investigation. Fe atoms contribute additional structural stability.

A detailed examination of FeMnScGa's electronic structure highlighted unique properties that complement those of other studied compounds. The observed spin-down bandgap of 0.442 eV (mBJ) falls within an intermediate range, potentially providing an ideal compromise between spin polarization and thermal stability. Following the Slater-Pauling rule, the material maintains a total magnetic moment of 3 μ_B . A thorough investigation of magnetic interactions highlights Mn atoms as the primary contributors, while Fe atoms exhibit minor positive moments and Sc and Ga atoms induce small

negative moments, forming a stable magnetic structure well-suited for spintronic applications.

4.3 Optical Properties and Light-Matter Interactions

FeMnCrGe's distinctive optical characteristics, revealed through examination, make it a strong candidate for diverse optoelectronic applications. The material exhibits low reflectivity in both visible and ultraviolet spectra. Solar energy conversion and UV detection are potential applications due to this efficient electromagnetic wave interaction. Furthermore, its spin-dependent optical properties expand its potential in hybrid spintronic-photonic systems. FeMnScAl exhibits prominent ultraviolet absorption peaks and exceptional optical conductivity. Along with its spin-dependent reflectivity and effective charge carrier generation, these properties point to the material's potential in cutting-edge optoelectronic applications. The material's distinct polarization effects and spin-dependent optical characteristics open opportunities for the design and implementation of spin-controlled devices. Moreover, significant plasmonic resonances were observed in the energy loss function; therefore, practical uses in plasmonic technologies and sensors are suggested. UV absorption coefficients exceeding $180 \times 10^4/\text{cm}$, along with significant interband transitions, contribute to FeMnScGa's unique optical properties. Due to its high refractive index in the visible and infrared regions, strong dielectric response, and effective light-matter interaction, the material stands out as a leading option for photovoltaic devices. The observed robust collective plasmon excitations suggest potential applications in plasmonic devices and optical sensing technologies.

4.4 Thermoelectric Properties and Energy Conversion

The study of thermoelectric properties revealed encouraging results, suggesting strong potential for applications in energy conversion. FeMnCrGe's spin-down channel showed outstanding thermoelectric efficiency, which achieved a Seebeck coefficient of about $124.1 \mu\text{V/K}$ at 500 K and a ZT value of 0.42. The power factor of $1.6 \times 10^{11} (\Omega \text{ m s})^{-1}$ at 500 K highlights the material's potential for thermal energy harvesting. High-temperature thermoelectric devices would benefit from this material's p-type charge carrier dominance and enhanced high-temperature performance.

FeMnScAl achieved an impressive peak ZT value of 0.689 at 150 K under the mBJ approximation, with a room temperature Seebeck coefficient of $196 \mu\text{V/K}$ demonstrating excellent thermoelectric response. The material's n-type conduction across the temperature range and decreasing lattice thermal conductivity with temperature suggest optimal conditions for thermoelectric energy conversion. The enhanced power factor at higher temperatures indicates particular promise for waste heat recovery applications.

Analysis of FeMnScGa's thermoelectric properties across a broad temperature range of 50-600 K revealed intricate temperature-dependent behavior. At 600 K (mBJ), the spin-up channel recorded a ZT value of 0.06094, with a corresponding power factor of $4.58 \times 10^{11} \text{ W/mK}^2$. The notable decline in lattice thermal conductivity from 1.93 to 0.161 W/mK^2 as temperature increased implies superior thermoelectric performance at higher temperatures. With dominant p-type carriers and efficient thermal-to-electrical energy conversion, the material shows promise for use in high-temperature thermoelectric devices.

4.5 Technological Implications and Applications

The study results have widespread applications in a number of technological fields. They are found to be highly spin polarized. The higher rate of spin polarization coupled with the in-built stable magnetic nature of these compounds offers the best condition for efficient spin injection and ensures consistent output in spintronic devices. Half-metallicity and spin-dependent transport make these materials excellent for a variety of spintronic applications. From magnetic tunnel junctions storing data to spin valve devices detecting magnetic fields, from MRAM components to spin-based logic circuits and spin-filter devices generating spin-polarized currents, the potential uses for these materials span a broad range.

The thermoelectric properties of these materials make them worth consideration in waste heat recovery and thermal energy harvesting in energy systems. Identified optical properties are relevant to future uses in photovoltaics for solar energy capture, ultraviolet photodetection for environmental monitoring, optical sensing for industrial applications, light-matter enhancement through plasmonic devices, and in energy-efficient light and display applications. In addition to these uses, these materials show

great versatility across multiple uses, highlighting their tremendous potential for spintronics and energy applications.

4.6 Scientific Impact and Methodological Contributions

Advancing the theoretical foundation of quaternary Heusler alloys, this research has provided critical insights, especially concerning FeMnCrGe, FeMnScAl, and FeMnScGa. This comprehensive analysis has provided crucial insights into several key aspects, including the structure-property relationships in complex alloy systems, the magnetic interaction mechanisms present in multi-component materials, and the engineering of electronic structures to optimize functionality. Furthermore, the study explores optimization strategies for material properties and offers predictive capabilities for material design.

This study has provided substantial contributions to materials science through the development and validation of key methodological approaches. These frameworks, created using the recommended methods, assist in accurate electronic structure calculations, the prediction of magnetic properties, the analysis of thermoelectric performance, the assessment of optical characteristics, and the evaluation of mechanical and dynamical stability.

4.7 Current Limitations and Technical Challenges

In a broad range of applications, Density Functional Theory (DFT) has proven highly effective due to its ability to provide fundamental insights into material and molecular properties. The accuracy of its results is affected by significant challenges, including delocalization and static correlation errors. DFT, although a powerful tool, is constrained by delocalization and static correlation errors, which compromise its predictive accuracy. These errors distort the calculation of key properties such as reaction barriers, band gaps, and charge transfer energies. As a result, DFT's efficacy in modeling complex systems is reduced.

The fundamental problem lies in the approximations inherent in the exchange-correlation functional of DFT. A significant boost to DFT's reliability, and a broadening of its impact in materials science and theoretical chemistry, could be achieved by designing improved functionals that address these limitations.

Experimental validation is critical, as the limited experimental data available for these alloys calls for further synthesis and characterization to validate computational results. The study assumes perfect crystal structures and the absence of defects as its framework. Impurities and degradation often cause real-world materials to deviate from this ideal, altering their performance.

4.8 Future Research Directions and Recommendations

The findings and limitations of this work highlight several promising directions for future research efforts focused on these three QHA.

4.8.1 Experimental Studies

The methods used in processing different materials need to be formulated with great care to ensure consistency and control of the composition of materials. Processing of large samples is done with optimal efficiency through methods like arc melting and solid-state reactions with exact stoichiometric studies and improving processing conditions to ensure exact composition control. Such processing methods provide for easy development of materials with higher levels of purity and make them amenable to advanced characterization and detailed studies.

The resulting compounds will be compared based on their structural properties using X-ray diffraction (XRD), which is also a key tool for identifying phase formation. Atomic-scale properties will be investigated using transmission electron microscopy (TEM), while surface morphology inspection will be carried out using scanning electron microscopy (SEM). In addition, our magnetic properties study—both those that arise in relation to phase transition and those that are magnetic in nature—will be complemented by methods like vibrating sample magnetometry (VSM) and superconducting quantum interference devices (SQUID).

The electronic and optical properties require experimental proof, and for this to happen, advanced characterization methods are required. In a detailed study of the electronic structure and surface states, X-ray photoelectron spectroscopy (XPS), scanning tunneling microscopy (STM), and angle-resolved photoemission spectroscopy (ARPES) can be used. To accurately determine excitonic excitations and band gaps, UV-LV and photoluminescence (PL) spectroscopy are necessary tools.

Evaluating thermoelectric materials requires precise measurements of key parameters. These include the Seebeck coefficient, electrical resistivity, thermal conductivity, and the Figure of Merit (ZT). Both the suitability assessment and efficiency optimization of thermoelectric materials, across a range of operating conditions, hinge on the precision of these measurements.

In order to be useful, materials should be fabricated into ultrathin films using techniques like sputtering or laser deposition using pulses (PLD). After fabrication, these films can be incorporated into useful devices, for which their function has to be tested to establish their feasibility for application in electronics, spintronics, or thermo-electronics.

4.8.2 Computational Studies

Quaternary Heusler Alloys (QHA) properties can be analyzed with various computation software packages, not only WIEN2k, since each of these packages has its own unique strengths that enhance the validity of results. Vienna Ab initio Simulation Package (VASP) is well known for its comprehensive density functional theory (DFT) features, demonstrating proficiency in electronic structure calculations, structural relaxations, and molecular dynamics simulations. Known to be very flexible, Quantum ESPRESSO, which is an open-source DFT software suite, has strong support for a variety of exchange-correlation functionals and is well-suited for surface, interface, and defect studies. Another prominent open-source code, ABINIT, is known to possess very rich electronic structure analysis capabilities and support for a wide variety of pseudopotentials. CASTEP, which is a commercial DFT code, is renowned for its user-friendliness and excels in the computation of mechanical, thermal, and optical properties. SIESTA is, however, excellent at treating large systems, such as biomolecules and nanostructures, with localized basis sets for improved computational efficiency.

To successfully achieve research objectives, the choice of computational software must be made on the basis of the requirements of the system, desired accuracy, and computer resources available.

4.9 Concluding Remarks and Future Outlook

A comprehensive study of the Heusler quaternary alloys FeMnCrGe, FeMnScAl, and FeMnScGa has revealed their outstanding potential, thereby highlighting their significance in advancing electronic and energy conversion technologies. The unique integration of half-metallicity, robust magnetic properties, and outstanding thermoelectric properties designates their critical role as pivotal materials for emerging developments. The findings in this work provide a strong foundation for stimulating further experimental investigations and practical applications. The manifested features indicate that such materials may have a key role in solving existing technological problems in the fields of spintronics, energy conversion, and light applications. Their excellent performance characteristics, especially regarding spin polarization, thermoelectric performance, and optical response, are very promising for practical applications. The theoretical basis and methodological developments established in this work offer a firm ground for material science, allowing follow-up research and innovation in analogous material systems. As technology continues to evolve and demand for even more efficient and enhanced materials continues to rise, the findings presented in this thesis continue to grow in relevance. The research identifies the constraints encountered and suggests potential avenues for future research, thus providing useful information for future studies and allowing the further development of these three materials. Their potential use is larger than their present applications, indicating the prospect of revolutionary advances in electronic devices, energy conversion devices, and other fields. Such a material's high potential offers a very wide scope for enhancement and use in high-tech applications. While experimental and theoretical studies are ongoing, the establishment of new properties and applications yet to be discovered will further assert the fundamental role of quaternary Heusler alloys in advancing technological advancements in the years to come. Their adaptability and excellent properties point to their usefulness in material research, indicating enormous potential in diverse fields of application.

List of Abbreviations

Abbreviation	Meaning	Description
<i>DFT</i>	Density Functional Theory	A quantum mechanical approach employed to study the electronic structure of systems with many interacting particles.
<i>QHA</i>	Quaternary Heusler Alloy	Comprising four distinct elements, this alloy is specially studied for its one-of-a-kind properties.
<i>FP-LAPW</i>	Full-Potential Linearized Augmented Plane Wave	An exceptionally precise computational method employed to determine the electronic properties of materials.
<i>GGA</i>	Generalized Gradient Approximation	A method within DFT that enhances calculation accuracy by incorporating the gradient of the electron density.
<i>mBJ</i>	Modified Becke-Johnson	A method used to improve the prediction of band gaps in semiconductors and insulators.
<i>ZT</i>	Figure of Merit (thermoelectric performance measure)	A dimensionless number that indicates the efficiency of a thermoelectric material.
μB	Bohr Magnetron (unit of magnetic moment)	A physical constant used to express the magnetic moment of particles, atoms, and molecules.
<i>eV</i>	Electron Volt (unit of energy)	This energy measure is a standard reference in atomic and particle physics.
<i>UV</i>	Ultraviolet	A category of electromagnetic radiation defined by its shorter wavelengths compared to visible light.
<i>R</i>	Reflectivity	A surface's ability to reflect light or radiation is quantified using this proportion.
$\sigma_1(\omega)$	Real part of the optical conductivity spectrum	The real portion of the optical conductivity provides insight into how efficiently a material conducts electricity in response to an optical field.
$\sigma_2(\omega)$	Imaginary part of the optical conductivity spectrum	The imaginary component of the optical conductivity, which corresponds to the energy dissipation within the material.
<i>B</i>	Bulk Modulus	This metric assesses a material's capacity to withstand uniform compression.
E_{Form}	Formation Energy	The energy change accompanying the formation of a compound from its elements under standard conditions is a critical measure.
a_0	Lattice Parameter	This refers to the measurable dimensions of the unit cell in a crystal lattice structure.

V_0	Equilibrium Volume	This refers to the volume occupied by the unit cell when the system is in equilibrium.
B'	Pressure Derivative of the Bulk Modulus	The bulk modulus variation with pressure is illustrated, offering valuable insights into how materials behave when subjected to stress.
DOS	Density of States	A function that specifies the quantity of states available for occupation at every energy level.

References

- [1] C. Felser, G.H. Fecher, B. Balke, Spintronics: A Challenge for Materials Science and Solid-State Chemistry, *Angewandte Chemie International Edition* 46 (2007) 668–699. <https://doi.org/10.1002/ANIE.200601815>.
- [2] L. Huang, R. He, S. Chen, H. Zhang, K. Dahal, H. Zhou, H. Wang, Q. Zhang, Z. Ren, A new n-type half-Heusler thermoelectric material NbCoSb, *Mater Res Bull* 70 (2015) 773–778. <https://doi.org/10.1016/J.MATERRESBULL.2015.06.022>.
- [3] V. Alijani, J. Winterlik, G.H. Fecher, S.S. Naghavi, S. Chadov, T. Gruhn, C. Felser, Quaternary Heusler compounds $\text{Co}_{2-x}\text{Rh}_x\text{MnZ}$ ($Z = \text{Ga}, \text{Sn}, \text{Sb}$): crystal structure, electronic structure, and magnetic properties, *Journal of Physics: Condensed Matter* 24 (2012) 046001. <https://doi.org/10.1088/0953-8984/24/4/046001>.
- [4] L. Bainsla, K.Z. Suzuki, M. Tsujikawa, H. Tsuchiura, M. Shirai, S. Mizukami, Magnetic tunnel junctions with an equiatomic quaternary CoFeMnSi Heusler alloy electrode, *Appl Phys Lett* 112 (2018). <https://doi.org/10.1063/1.5002763>.
- [5] X. Lu, D.T. Morelli, Y. Xia, F. Zhou, V. Ozolins, H. Chi, X. Zhou, C. Uher, High Performance Thermoelectricity in Earth-Abundant Compounds Based on Natural Mineral Tetrahedrites, *Adv Energy Mater* 3 (2013) 342–348. <https://doi.org/10.1002/aenm.201200650>.
- [6] L. Bainsla, R. Yilgin, J. Okabayashi, A. Ono, K. Suzuki, S. Mizukami, Structural and magnetic properties of epitaxial thin films of the equiatomic quaternary CoFeMnSi Heusler alloy, *Phys Rev B* 96 (2017) 094404. <https://doi.org/10.1103/PhysRevB.96.094404>.
- [7] V. Alijani, O. Meshcheriakova, J. Winterlik, G. Kreiner, G.H. Fecher, C. Felser, Increasing Curie temperature in tetragonal Mn_2RhSn Heusler compound through substitution of Rh by Co and Mn by Rh, *J Appl Phys* 113 (2013). <https://doi.org/10.1063/1.4791564>.
- [8] R. Prakash, G. Kalpana, First-principles study on novel Fe-based quaternary Heusler alloys, with robust half-metallic, thermoelectric and optical properties, *RSC Adv* 13 (2023) 10847–10860. <https://doi.org/10.1039/D3RA00942D>.
- [9] K. Chinnadurai, B. Natesan, First principles calculations of 3d-4d transition metal based LiMgPdSn-type FeCrRuZ ($Z = \text{Al}, \text{Ga}, \text{In}, \text{Si}$) equiatomic quaternary Heusler alloys, *Comput Mater Sci* 188 (2021). <https://doi.org/10.1016/j.commatsci.2020.110116>.
- [10] S. Idrissi, S. Ziti, H. Labrim, L. Bahmad, I. El Housni, R. Khalladi, S. Mtougui, N. El Mekkaoui, Half-metallic behavior and magnetic proprieties of the quaternary Heusler alloys YFeCrZ ($Z = \text{Al}, \text{Sb}$ and Sn), *J Alloys Compd* 820 (2020) 153373. <https://doi.org/10.1016/J.JALLCOM.2019.153373>.
- [11] M. Ketfi, S. Berri, D. Maouche, N. Bouarissa, Structural, Electronic, Half-Metallic and Thermoelectric Properties of Quaternary Heusler Alloys AgCoFeZ

- ($Z = \text{Al, Ga, Si, Ge, and Sn}$), NiFeCrZ ($Z = \text{Al, Si, Ge, In}$) and NdCoMnGa : a First Principles Study, *J Supercond Nov Magn* 37 (2024) 737–751. <https://doi.org/10.1007/s10948-024-06701-0>.
- [12] Z. Xia, Z. Liu, Q. Zhang, Y. Zhang, X. Ma, Magnetic and anomalous transport properties in spin-gapless semiconductor like quaternary Heusler alloy CoFeTiSn , *J Magn Magn Mater* 553 (2022) 169283. <https://doi.org/10.1016/j.jmmm.2022.169283>.
- [13] X.-P. Wei, Z.-Y. Mei, X. Liu, X. Tao, Investigations on stability, Gilbert damping, electronic and magnetic properties along with Curie temperature for quaternary Heusler alloys CrTiCoZ , *Results Phys* 58 (2024) 107512. <https://doi.org/10.1016/j.rinp.2024.107512>.
- [14] Z. Khadhraoui, S.E. Amara, A.Y. Behlali, H. Kheribot, S. Makhlouf, S. Labidi, A. Amara, Comprehensive analysis: Exploring quaternary Heusler alloys CoFeXGe ($X = \text{Hf and Ta}$) through first-principles calculations, *Journal of the American Ceramic Society* 107 (2024) 7421–7440. <https://doi.org/10.1111/jace.20014>.
- [15] J. He, S.S. Naghavi, V.I. Hegde, M. Amsler, C. Wolverton, Designing and Discovering a New Family of Semiconducting Quaternary Heusler Compounds Based on the 18-Electron Rule, *Chemistry of Materials* 30 (2018) 4978–4985. <https://doi.org/10.1021/acs.chemmater.8b01096>.
- [16] D. Rani, L. Bainsla, K.G. Suresh, A. Alam, Experimental and theoretical investigation on the possible half-metallic behaviour of equiatomic quaternary Heusler alloys: CoRuMnGe and CoRuVZ ($Z = \text{Al, Ga}$), *J Magn Magn Mater* 492 (2019) 165662. <https://doi.org/10.1016/j.jmmm.2019.165662>.
- [17] H. Frioui, A. Meddour, M.H. Gous, Structural, mechanical, electronic, magnetic, optical, and thermoelectric properties of a new equiatomic quaternary heusler compound FeZrCrZ ($Z = \text{Si, Ge, Sn}$): A first-principles investigation, *Mater Sci Semicond Process* 182 (2024) 108697. <https://doi.org/10.1016/j.mssp.2024.108697>.
- [18] L. Suktel, S.M. Saini, Thermoelectric performance of newly discovered LiHfPdZ ($Z = \text{Al, Ga, and In}$) quaternary Heusler compounds: A DFT study, *Physica B Condens Matter* 690 (2024) 416268. <https://doi.org/10.1016/j.physb.2024.416268>.
- [19] R. Guo, G. Liu, X. Wang, H. Rozale, L. Wang, R. Khenata, Z. Wu, X. Dai, First-principles study on quaternary Heusler compounds ZrFeVZ ($Z = \text{Al, Ga, In}$) with large spin-flip gap, *RSC Adv* 6 (2016) 109394–109400. <https://doi.org/10.1039/C6RA18873G>.
- [20] A.A. Mubarak, S. Saad, F. Hamioud, M. Al-Elaimi, Structural, thermo-elastic, electro-magnetic and thermoelectric attributes of quaternary CoNbMnX ($X = \text{Al, Si}$) Heusler alloys, *Solid State Sci* 111 (2021) 106397. <https://doi.org/10.1016/j.solidstatesciences.2020.106397>.

- [21] N.O. Nenuwe, S.E. Kpuwhara, Physical properties of CoX'ZrGa (X' = V, Cr) quaternary-Heuslers: A first-principles investigation, *Physica B Condens Matter* 685 (2024) 416002. <https://doi.org/10.1016/j.physb.2024.416002>.
- [22] A. Fert, Nobel Lecture: Origin, development, and future of spintronics, *Rev Mod Phys* 80 (2008) 1517–1530. <https://doi.org/10.1103/RevModPhys.80.1517>.
- [23] L.B. Chandrasekar, K. Gnanasekar, M. Karunakaran, Spintronics – A mini review, *Superlattices Microstruct* 136 (2019) 106322. <https://doi.org/10.1016/j.spmi.2019.106322>.
- [24] G.G. Baez Flores, M. van Schilfgaarde, K.D. Belashchenko, Tunneling magnetoresistance in MgO tunnel junctions with Fe-based leads in empirically corrected density functional theory, *Phys Rev B* 110 (2024) 224425. <https://doi.org/10.1103/PhysRevB.110.224425>.
- [25] O. Wessely, B. Skubic, L. Nordström, Spin-transfer torque in helical spin-density waves, *Phys Rev B* 79 (2009) 104433. <https://doi.org/10.1103/PhysRevB.79.104433>.
- [26] F. Birch, Finite Elastic Strain of Cubic Crystals, *Physical Review* 71 (1947) 809–824. <https://doi.org/10.1103/PhysRev.71.809>.
- [27] V.G. Tyuterev, N. Vast, Murnaghan's equation of state for the electronic ground state energy, *Comput Mater Sci* 38 (2006) 350–353. <https://doi.org/10.1016/J.COMMATSCI.2005.08.012>.
- [28] F.D. Murnaghan, The Compressibility of Media under Extreme Pressures, *Proceedings of the National Academy of Sciences* 30 (1944) 244–247. <https://doi.org/10.1073/pnas.30.9.244>.
- [29] S.M. Azar, A.A. Mousa, J.M. Khalifeh, Structural, electronic and magnetic properties of Ti_{1+x}FeSb Heusler alloys, *Intermetallics (Barking)* 85 (2017) 197–205. <https://doi.org/10.1016/J.INTERMET.2017.02.018>.
- [30] Z. Zeng, F. Calle-Vallejo, M.B. Mogensen, J. Rossmeisl, Generalized trends in the formation energies of perovskite oxides, *Physical Chemistry Chemical Physics* 15 (2013) 7526–7533. <https://doi.org/10.1039/C3CP50257K>.
- [31] V. Stevanović, S. Lany, X. Zhang, A. Zunger, Correcting density functional theory for accurate predictions of compound enthalpies of formation: Fitted elemental-phase reference energies, *Phys Rev B* 85 (2012) 115104. <https://doi.org/10.1103/PhysRevB.85.115104>.
- [32] J. Zhu, F. Wang, B. Wang, Y. Wang, J. Liu, W. Zhang, Z. Wen, Surface Acidity as Descriptor of Catalytic Activity for Oxygen Evolution Reaction in Li-O₂ Battery, *J Am Chem Soc* 137 (2015) 13572–13579. <https://doi.org/10.1021/jacs.5b07792>.

- [33] A. Wang, Y. Yin, Z. Sun, G. Jin, C. Xin, The Application of Supervised Learning Algorithms in Predicting the Formation Energy of NLO Crystals, *Adv Theory Simul* 7 (2024). <https://doi.org/10.1002/adts.202400048>.
- [34] Y.K. Rajnath, S. Tiwari, V. Kumar, Thermodynamics for Mechatronics, in: *Computational Intelligent Techniques in Mechatronics*, Wiley, 2024: pp. 41–81. <https://doi.org/10.1002/9781394175437.ch2>.
- [35] F. Mouhat, F.-X. Coudert, Necessary and sufficient elastic stability conditions in various crystal systems, *Phys Rev B* 90 (2014) 224104. <https://doi.org/10.1103/PhysRevB.90.224104>.
- [36] R. Golesorkhtabar, P. Pavone, J. Spitaler, P. Puschnig, C. Draxl, ElaStic: A tool for calculating second-order elastic constants from first principles, *Comput Phys Commun* 184 (2013) 1861–1873. <https://doi.org/10.1016/j.cpc.2013.03.010>.
- [37] C.M. Zener, S. Siegel, Elasticity and Anelasticity of Metals., *J Phys Colloid Chem* 53 (1949) 1468. <https://doi.org/10.1021/j150474a017>.
- [38] I.N., V.F.F. and B.S.A., I.N. Frantsevich, Elastic constants and elastic moduli of metals and insulators (In Russian). *Izdatel'stvo Naukova Dumka*, Kiev, Reference Book (1982) 280.
- [39] M. Jamal, Cubic-elastic, [http, WIEN2k. at/Reg_user/Unsupported/Cubic-Elast](http://WIEN2k.at/Reg_user/Unsupported/Cubic-Elast) (2012).
- [40] P.H. Mott, J.R. Dorgan, C.M. Roland, The bulk modulus and Poisson's ratio of "incompressible" materials, *J Sound Vib* 312 (2008) 572–575. <https://doi.org/10.1016/j.jsv.2008.01.026>.
- [41] N.K. Mukhopadhyay, P. Paufler, Micro- and nanoindentation techniques for mechanical characterisation of materials, *International Materials Reviews* 51 (2006) 209–245. <https://doi.org/10.1179/174328006X102475>.
- [42] H.J. McSkimin, P. Andreatch, Elastic Moduli of Diamond as a Function of Pressure and Temperature, *J Appl Phys* 43 (1972) 2944–2948. <https://doi.org/10.1063/1.1661636>.
- [43] K.K. Phani, D. Sanyal, The relations between the shear modulus, the bulk modulus and Young's modulus for porous isotropic ceramic materials, *Materials Science and Engineering: A* 490 (2008) 305–312. <https://doi.org/10.1016/j.msea.2008.01.030>.
- [44] J.W. Soh, H.M. Lee, H.S. Kwon, Relation between Poisson's ratio and ionicity in simple binary cubic compounds, *J Alloys Compd* 194 (1993) 119–125. [https://doi.org/10.1016/0925-8388\(93\)90656-8](https://doi.org/10.1016/0925-8388(93)90656-8).
- [45] L.J. Zukor, Nature and properties of engineering materials. Z. D. Jastrzebski. Wiley, New York- London, 1959, xvii + 571 pp. \$11.00, *J Appl Polym Sci* 4 (1960) 372–373. <https://doi.org/10.1002/app.1960.070041224>.

- [46] E. Rybacki, A. Reinicke, T. Meier, M. Makasi, G. Dresen, What controls the mechanical properties of shale rocks? – Part I: Strength and Young’s modulus, *J Pet Sci Eng* 135 (2015) 702–722. <https://doi.org/10.1016/j.petrol.2015.10.028>.
- [47] P. Ravindran, L. Fast, P.A. Korzhavyi, B. Johansson, J. Wills, O. Eriksson, Density functional theory for calculation of elastic properties of orthorhombic crystals: Application to TiSi₂, *J Appl Phys* 84 (1998) 4891–4904. <https://doi.org/10.1063/1.368733>.
- [48] D. Jiang, M. Wu, D. Liu, F. Li, M. Chai, S. Liu, Structural Stability, Electronic Structures, Mechanical Properties and Debye Temperature of Transition Metal Impurities in Tungsten: A First-Principles Study, *Metals (Basel)* 9 (2019) 967. <https://doi.org/10.3390/met9090967>.
- [49] C. Kittel, P. McEuen, Introduction to solid state physics, John Wiley & Sons, 2018.
- [50] Solid State Physics. Von N. W. Ashcroft und N. D. Mermin; Holt, Rinehart and Winston, New York 1976, XXII, 826 Seiten, \$19,95, *Physik in Unserer Zeit* 9 (1978) 33–33. <https://doi.org/10.1002/piuz.19780090109>.
- [51] S.H. Simon, The Oxford solid state basics, OUP Oxford, 2013.
- [52] P.Y. Yu, M. Cardona, Fundamentals of Semiconductors, Springer Berlin Heidelberg, Berlin, Heidelberg, 2010. <https://doi.org/10.1007/978-3-642-00710-1>.
- [53] N.M. Harrison, An introduction to density functional theory, *Nato Science Series Sub Series III Computer and Systems Sciences* 187 (2003) 45–70.
- [54] P. Hohenberg, W. Kohn, Inhomogeneous electron gas, *Physical Review* 136 (1964) B864. <https://doi.org/10.1103/PHYSREV.136.B864/FIGURE/1/THUMB>.
- [55] W. Kohn, others, W. kohn and Ij sham, *phys. rev.* 140, a1133 (1965), *Phys. Rev* 140 (1965) A1133.
- [56] M.P. Marder, Condensed matter physics, John Wiley & Sons, 2010.
- [57] J.F. Schenck, The role of magnetic susceptibility in magnetic resonance imaging: MRI magnetic compatibility of the first and second kinds, *Med Phys* 23 (1996) 815–850. <https://doi.org/10.1118/1.597854>.
- [58] M. Grimsditch, A. Hoffmann, P. Vavassori, H. Shi, D. Lederman, Exchange-Induced Anisotropies at Ferromagnetic-Antiferromagnetic Interfaces above and below the Néel Temperature, *Phys Rev Lett* 90 (2003) 257201. <https://doi.org/10.1103/PhysRevLett.90.257201>.
- [59] D. Ghernaout, Magnetic field generation in the water treatment perspectives: An overview, *International Journal of ADVANCED AND APPLIED SCIENCES* 5 (2018) 193–203. <https://doi.org/10.21833/ijaas.2018.01.025>.
- [60] I. Galanakis, Slater–Pauling Behavior in Half-Metallic Heusler Compounds, *Nanomaterials* 13 (2023) 2010. <https://doi.org/10.3390/nano13132010>.

- [61] A. FUJISHIMA, K. HONDA, Electrochemical Photolysis of Water at a Semiconductor Electrode, *Nature* 238 (1972) 37–38. <https://doi.org/10.1038/238037a0>.
- [62] S.O. Kasap, *Electronic materials and Devices*, McGraw-Hill New York, 2006.
- [63] J.A. Schuller, E.S. Barnard, W. Cai, Y.C. Jun, J.S. White, M.L. Brongersma, Plasmonics for extreme light concentration and manipulation, *Nat Mater* 9 (2010) 193–204. <https://doi.org/10.1038/nmat2630>.
- [64] T. Lantri, S. Bentata, B. Bouadjemi, W. Benstaali, B. Bouhafs, A. Abbad, A. Zitouni, Effect of Coulomb interactions and Hartree-Fock exchange on structural, elastic, optoelectronic and magnetic properties of Co₂MnSi Heusler: A comparative study, *J Magn Magn Mater* 419 (2016) 74–83. <https://doi.org/10.1016/j.jmmm.2016.06.012>.
- [65] M.S. Dresselhaus, *Solid State Physics Optical Properties of Solids*, (2022).
- [66] M. Fox, *Optical properties of solids*, Oxford university press, 2010.
- [67] S.A. Khan, A.H. Reshak, Z.A. Alahmed, Electronic band structure and optoelectronic properties of SrCu₂X₂ (X = As, Sb): DFT calculation, *J Mater Sci* 49 (2014) 5208–5217. <https://doi.org/10.1007/S10853-014-8230-3>/METRICS.
- [68] H. Zaari, M. Boujnah, A. El Hachimi, A. Benyoussef, A. El Kenz, Optical properties of ZnTe doped with transition metals (Ti, Cr and Mn), *Opt Quantum Electron* 46 (2014) 75–86. <https://doi.org/10.1007/s11082-013-9708-y>.
- [69] X. Leng, F. Jin, M. Wei, Y. Ma, <sc>GW</sc> method and Bethe–Salpeter equation for calculating electronic excitations, *WIREs Computational Molecular Science* 6 (2016) 532–550. <https://doi.org/10.1002/wcms.1265>.
- [70] T.M. Bhat, D.C. Gupta, Robust thermoelectric performance and high spin polarisation in CoMnTiAl and FeMnTiAl compounds, *RSC Adv* 6 (2016) 80302–80309. <https://doi.org/10.1039/C6RA18934B>.
- [71] D.C. SPANNER, The Peltier Effect and its Use in the Measurement of Suction Pressure, *J Exp Bot* 2 (1951) 145–168. <https://doi.org/10.1093/jxb/2.2.145>.
- [72] C. Wood, Materials for thermoelectric energy conversion, *Reports on Progress in Physics* 51 (1988) 459. <https://doi.org/10.1088/0034-4885/51/4/001>.
- [73] S.B. Riffat, X. Ma, Thermoelectrics: a review of present and potential applications, *Appl Therm Eng* 23 (2003) 913–935. [https://doi.org/10.1016/S1359-4311\(03\)00012-7](https://doi.org/10.1016/S1359-4311(03)00012-7).
- [74] C.L. Hapenciuc, T. Borca-Tasciuc, I.N. Mihailescu, The relationship between the thermoelectric generator efficiency and the device engineering figure of merit $Z_{d,eng}$. The maximum efficiency η_{max} , *AIP Adv* 7 (2017). <https://doi.org/10.1063/1.4979328>.
- [75] A. Bejan, A.D. Kraus, *Heat transfer handbook*, John Wiley & Sons, 2003.

- [76] S. Singh, R.K. Maurya, S.K. Pandey, Investigation of thermoelectric properties of ZnV_2O_4 compound at high temperatures, *J Phys D Appl Phys* 49 (2016) 425601. <https://doi.org/10.1088/0022-3727/49/42/425601>.
- [77] M.-K. Han, K. Hoang, H. Kong, R. Pcionek, C. Uher, K.M. Paraskevopoulos, S.D. Mahanti, M.G. Kanatzidis, Substitution of Bi for Sb and its Role in the Thermoelectric Properties and Nanostructuring in $\text{Ag}_{1-x}\text{Pb}_{18}\text{MTe}_{20}$ ($\text{M} = \text{Bi}, \text{Sb}$) ($x = 0, 0.14, 0.3$), *Chemistry of Materials* 20 (2008) 3512–3520. <https://doi.org/10.1021/cm703661g>.
- [78] H. Alqurashi, R. Haleoot, A. Pandit, B. Hamad, Investigations of the electronic, dynamical, and thermoelectric properties of $\text{Cd}_{1-x}\text{Zn}_x\text{O}$ alloys: First-principles calculations, *Mater Today Commun* 28 (2021) 102511. <https://doi.org/10.1016/J.MTCOMM.2021.102511>.
- [79] X.-L. Shi, J. Zou, Z.-G. Chen, Advanced Thermoelectric Design: From Materials and Structures to Devices, *Chem Rev* 120 (2020) 7399–7515. <https://doi.org/10.1021/acs.chemrev.0c00026>.
- [80] Z. Zada, A.A. Khan, A.H. Reshak, I. Khan, S. Zada, M. Ismail, M. Fazal-ur-Rehman, M. Saqib, G. Murtaza, Q. Khan, M.M. Ramli, Insight into the electronic structure, magnetic, and thermoelectric properties of transition metal pnictides KCr_2L_2 ($\text{K} = \text{Ca}, \text{Sr}; \text{L} = \text{P}, \text{As}$): As substitute source for renewing energy, *Physica B Condens Matter* 649 (2023) 414470. <https://doi.org/10.1016/j.physb.2022.414470>.
- [81] Z. Zada, R. Zada, A.A. Khan, M. Saqib, M.F.U. Rehman, M. Ismail, N. Kulhari, K.S. Sharma, M. Ismail, X. Shen, M. Faizan, Investigation of electronic structure, magnetic stability, spin coupling, and thermodynamic properties of novel antiferromagnets XMn_2Y_2 ($\text{X} = \text{Ca}, \text{Sr}; \text{Y} = \text{P}, \text{As}$), *J Mol Struct* 1268 (2022) 133698. <https://doi.org/10.1016/j.molstruc.2022.133698>.
- [82] A.A. Khan, Z. Zada, A.H. Reshak, M. Ishaq, S. Zada, M. Saqib, M. Ismail, M. Fazal-ur-Rehman, G. Murtaza, S. Zada, M.M. Ramli, GGA and GGA + U Study of ThMn_2Si_2 and ThMn_2Ge_2 Compounds in a Body-Centered Tetragonal Ferromagnetic Phase, *Molecules* 27 (2022) 7070. <https://doi.org/10.3390/molecules27207070>.
- [83] J. Sapirstein, Theoretical methods for the relativistic atomic many-body problem, *Rev Mod Phys* 70 (1998) 55. <https://doi.org/10.1103/RevModPhys.70.55>.
- [84] M. Born, R. Oppenheimer, Zur Quantentheorie der Molekeln, *Ann Phys* 389 (1927) 457–484. <https://doi.org/10.1002/andp.19273892002>.
- [85] J.C. Slater, A Simplification of the Hartree-Fock Method, *Physical Review* 81 (1951) 385–390. <https://doi.org/10.1103/PhysRev.81.385>.
- [86] R.G. Parr, Density Functional Theory of Atoms and Molecules, in: *Horizons of Quantum Chemistry*, Springer Netherlands, Dordrecht, 1980: pp. 5–15. https://doi.org/10.1007/978-94-009-9027-2_2.

- [87] C.A. Jiménez-Hoyos, T.M. Henderson, T. Tsuchimochi, G.E. Scuseria, Projected hartree–fock theory, *J Chem Phys* 136 (2012).
- [88] D.S. Sholl, J.A. Steckel, *Density functional theory□: a practical introduction*, Wiley, 2009. <https://www.wiley.com/en-us/Density+Functional+Theory%3A+A+Practical+Introduction-p-9780470447710> (accessed April 4, 2024).
- [89] M. Foley, P.A. Madden, Further orbital-free kinetic-energy functionals for *ab initio* molecular dynamics, *Phys Rev B* 53 (1996) 10589–10598. <https://doi.org/10.1103/PhysRevB.53.10589>.
- [90] A.Y. Shul'man, D. V Posvyanskii, Truly self-consistent solution of Kohn-Sham equations for extended systems with inhomogeneous electron gas, *J Phys Conf Ser* 510 (2014) 012029. <https://doi.org/10.1088/1742-6596/510/1/012029>.
- [91] M. Ernzerhof, G.E. Scuseria, Assessment of the Perdew–Burke–Ernzerhof exchange–correlation functional, *J Chem Phys* 110 (1999) 5029–5036. <https://doi.org/10.1063/1.478401>.
- [92] J.P. Perdew, K. Burke, M. Ernzerhof, Generalized Gradient Approximation Made Simple, *Phys Rev Lett* 77 (1996) 3865. <https://doi.org/10.1103/PhysRevLett.77.3865>.
- [93] D.M. Ceperley, B.J. Alder, Ground State of the Electron Gas by a Stochastic Method, *Phys Rev Lett* 45 (1980) 566–569. <https://doi.org/10.1103/PhysRevLett.45.566>.
- [94] J.A. Camargo-Martínez, R. Baquero, Performance of the modified Becke-Johnson potential for semiconductors, *Phys Rev B Condens Matter Mater Phys* 86 (2012) 195106. <https://doi.org/10.1103/PHYSREVB.86.195106>/FIGURES/2/MEDIUM.
- [95] F. Tran, WIEN2k: An Augmented Plane Wave Plus Local Orbitals Program for Calculating Crystal Properties, (2018).
- [96] P. Blaha, K. Schwarz, F. Tran, R. Laskowski, G.K.H. Madsen, L.D. Marks, WIEN2k: An APW+lo program for calculating the properties of solids, *Journal of Chemical Physics* 152 (2020). https://doi.org/10.1063/1.5143061/16727314/074101_1_ACCEPTED_MANUSCRIPT.PDF.
- [97] J.C. Slater, Wave Functions in a Periodic Potential, *Physical Review* 51 (1937) 846–851. <https://doi.org/10.1103/PhysRev.51.846>.
- [98] *Planewaves, Pseudopotentials and the LAPW Method*, Springer US, 2006. <https://doi.org/10.1007/978-0-387-29684-5>.
- [99] G.K.H. Madsen, D.J. Singh, BoltzTraP. A code for calculating band-structure dependent quantities, *Comput Phys Commun* 175 (2006) 67–71. <https://doi.org/10.1016/J.CPC.2006.03.007>.

- [100] D.T. Morelli, G.A. Slack, High Lattice Thermal Conductivity Solids, in: High Thermal Conductivity Materials, Springer-Verlag, New York, n.d.: pp. 37–68. https://doi.org/10.1007/0-387-25100-6_2.
- [101] M. Hariharan, E. R D, Investigations on the electronic, optical, thermoelectric and thermodynamic properties of quaternary coinage metal HgSeBr: a DFT study, Phys Scr 99 (2024) 035937. <https://doi.org/10.1088/1402-4896/ad26c0>.
- [102] T. Charpin, A package for calculating elastic tensors of cubic phase using WIEN, Laboratory of Geometrix, Paris (2001) 1–21.
- [103] S. Blügel, G. Bihlmayer, Full-potential linearized augmented planewave method, Computational Nanoscience: Do It Yourself 31 (2006) 85–129. <https://juser.fz-juelich.de/record/50026/files/FZJ-2014-02213.pdf> (accessed August 15, 2024).
- [104] P. Blaha, K. Schwarz, P. Sorantin, S.B. Trickey, Full-potential, linearized augmented plane wave programs for crystalline systems, Comput Phys Commun 59 (1990) 399–415. [https://doi.org/10.1016/0010-4655\(90\)90187-6](https://doi.org/10.1016/0010-4655(90)90187-6).
- [105] T.M. Bhat, S. Singh, D.C. Gupta, Electronic and Transport Properties of LaNi₄Sb₁₂ Skutterudite: Modified Becke–Johnson Approach, J Electron Mater 47 (2018) 4544–4549. <https://doi.org/10.1007/s11664-018-6323-5>.
- [106] F. Tran, P. Blaha, Accurate Band Gaps of Semiconductors and Insulators with a Semilocal Exchange–Correlation Potential, Phys Rev Lett 102 (2009) 226401. <https://doi.org/10.1103/PhysRevLett.102.226401>.
- [107] C. Ambrosch-Draxl, J.A. Majewski, P. Vogl, G. Leising, First-principles studies of the structural and optical properties of crystalline poly(*para* -phenylene), Phys Rev B 51 (1995) 9668–9676. <https://doi.org/10.1103/PhysRevB.51.9668>.
- [108] C. Ambrosch-Draxl, J.O. Sofo, Linear optical properties of solids within the full-potential linearized augmented planewave method, Comput Phys Commun 175 (2006) 1–14. <https://doi.org/10.1016/j.cpc.2006.03.005>.
- [109] H. Rathgen, M.I. Katsnelson, Symmetry Assumptions, Kramers-Kronig Transformation and Analytical Continuation in Ab Initio Calculations of Optical Conductivities, Phys Scr T109 (2004) 170. <https://doi.org/10.1238/Physica.Topical.109a00170>.
- [110] J. Wang, Y. Zhou, Dependence of elastic stiffness on electronic band structure of nanolaminate M₂A₁C (M=Ti,V,Nb, and Cr) ceramics, Phys Rev B Condens Matter Mater Phys 69 (2004) 214111. <https://doi.org/10.1103/PHYSREVB.69.214111/FIGURES/6/MEDIUM>.
- [111] Z. Zada, H. Ullah, R. Bibi, S. Zada, A. Mahmood, Electronic band profiles and magneto-electronic properties of ternary XCu₂P₂(X = Ca, Sr) compounds: Insight from ab initio calculations, Zeitschrift Fur Naturforschung - Section A Journal of Physical Sciences 75 (2020) 543–549. <https://doi.org/10.1515/ZNA-2020-0053/MACHINEREADABLECITATION/RIS>.

- [112] O. Gunnarsson, O.K. Andersen, O. Jepsen, J. Zaanen, Density-functional calculation of the parameters in the Anderson model: Application to Mn in CdTe, *Phys Rev B* 39 (1989) 1708. <https://doi.org/10.1103/PhysRevB.39.1708>.
- [113] A. Yakoubi, O. Baraka, B. Bouhafs, Structural and electronic properties of the Laves phase based on rare earth type BaM₂ (M = Rh, Pd, Pt), *Results Phys* 2 (2012) 58–65. <https://doi.org/10.1016/J.RINP.2012.06.001>.
- [114] Y. Toual, S. Mouchou, A. Azouaoui, A. Harbi, A. Hourmatallah, K. Bouslykhane, N. Benzakour, First principles study of structural and optoelectronic properties of semiconductor NaBeP_xN_{1-x} alloys for photovoltaic applications, *Comput Theor Chem* 1240 (2024) 114845. <https://doi.org/10.1016/j.comptc.2024.114845>.
- [115] K. Özdoğan, E. Şaşoğlu, I. Galanakis, Slater-Pauling behavior in LiMgPdSn-type multifunctional quaternary Heusler materials: Half-metallicity, spin-gapless and magnetic semiconductors, *J Appl Phys* 113 (2013). <https://doi.org/10.1063/1.4805063/370344>.
- [116] D.M. Hoat, M. Naseri, Examining the half-metallicity and thermoelectric properties of new equiatomic quaternary Heusler compound CoVRhGe under pressure, *Physica B Condens Matter* 583 (2020) 412058. <https://doi.org/10.1016/J.PHYSB.2020.412058>.
- [117] K. Bouferrache, M.A. Ghebouli, Y. Slimani, B. Ghebouli, M. Fatmi, T. Chihi, N. Algethami, S.A. Mouhammad, S. Alomairy, E.B. Elkenany, Structural stability, opto-electronic, magnetic and thermoelectric properties of half-metallic ferromagnets quaternary Heusler alloys CoFeXAs (X = Mn, Cr and V), *Solid State Commun* 377 (2024) 115366. <https://doi.org/10.1016/j.ssc.2023.115366>.
- [118] O. Lucia, P. Maussion, E.J. Dede, J.M. Burdio, Induction heating technology and its applications: Past developments, current technology, and future challenges, *IEEE Transactions on Industrial Electronics* 61 (2014) 2509–2520. <https://doi.org/10.1109/TIE.2013.2281162>.
- [119] M. Born, K. Huang, *Dynamical Theory Of Crystal Lattices*, Oxford University Press New York, NY, 1996. <https://doi.org/10.1093/oso/9780192670083.001.0001>.
- [120] S.F. Pugh, XCII. Relations between the elastic moduli and the plastic properties of polycrystalline pure metals, *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* 45 (1954) 823–843. <https://doi.org/10.1080/14786440808520496>.
- [121] O.L. Anderson, A simplified method for calculating the debye temperature from elastic constants, *Journal of Physics and Chemistry of Solids* 24 (1963) 909–917. [https://doi.org/10.1016/0022-3697\(63\)90067-2](https://doi.org/10.1016/0022-3697(63)90067-2).
- [122] D. V. Suetin, I.R. Shein, Electronic Structure, Mechanical and Dynamical Stability of Hexagonal Subcarbides M₂C (M = Tc, Ru, Rh, Pd, Re, Os, Ir, and Pt): Ab Initio Calculations, *Physics of the Solid State* 60 (2018) 213–224. <https://doi.org/10.1134/S1063783418020282>.

- [123] A.H. Reshak, Z.A. Alahmed, S. Azam, Electronic Structure, Electronic Charge Density and Optical Properties Analyses of $\text{Rb}_2\text{Al}_2\text{B}_2\text{O}_7$ Compound: DFT Calculation, *Int J Electrochem Sci* 9 (2014) 975–989. [https://doi.org/10.1016/S1452-3981\(23\)07771-4](https://doi.org/10.1016/S1452-3981(23)07771-4).
- [124] J.A. Abraham, G. Pagare, S.P. Sanyal, Electronic Structure, Electronic Charge Density, and Optical Properties Analysis of GdX_3 ($\text{X} = \text{In, Sn, Tl, and Pb}$) Compounds: DFT Calculations, *Indian Journal of Materials Science* 2015 (2015) 1–11. <https://doi.org/10.1155/2015/296095>.
- [125] T. Graf, C. Felser, S.S.P. Parkin, Simple rules for the understanding of Heusler compounds, *Progress in Solid State Chemistry* 39 (2011) 1–50. <https://doi.org/10.1016/J.PROGSOLIDSTCHEM.2011.02.001>.
- [126] S. V. Faleev, Y. Ferrante, J. Jeong, M.G. Samant, B. Jones, S.S.P. Parkin, Unified explanation of chemical ordering, the Slater-Pauling rule, and half-metallicity in full Heusler compounds, *Phys Rev B* 95 (2017) 045140. <https://doi.org/10.1103/PHYSREVB.95.045140/FIGURES/6/MEDIUM>.
- [127] Z. Zada, A.A. Khan, A.H. Reshak, I. Khan, S. Zada, M. Ismail, M. Fazal-ur-Rehman, M. Saqib, G. Murtaza, Q. Khan, M.M. Ramli, Insight into the electronic structure, magnetic, and thermoelectric properties of transition metal pnictides KCr_2L_2 ($\text{K} = \text{Ca, Sr; L} = \text{P, As}$): As substitute source for renewing energy, *Physica B Condens Matter* 649 (2023) 414470. <https://doi.org/10.1016/j.physb.2022.414470>.
- [128] Z. Zada, R. Zada, A.A. Khan, M. Saqib, M.F.U. Rehman, M. Ismail, N. Kulhari, K.S. Sharma, M. Ismail, X. Shen, M. Faizan, Investigation of electronic structure, magnetic stability, spin coupling, and thermodynamic properties of novel antiferromagnets XMn_2Y_2 ($\text{X} = \text{Ca, Sr; Y} = \text{P, As}$), *J Mol Struct* 1268 (2022) 133698. <https://doi.org/10.1016/j.molstruc.2022.133698>.
- [129] R. Loudon, The propagation of electromagnetic energy through an absorbing dielectric, *Journal of Physics A: General Physics* 3 (1970) 233–245. <https://doi.org/10.1088/0305-4470/3/3/008>.
- [130] J. Sun, H.-T. Wang, J. He, Y. Tian, Ab initio investigations of optical properties of the high-pressure phases of ZnO , *Phys Rev B* 71 (2005) 125132. <https://doi.org/10.1103/PhysRevB.71.125132>.
- [131] W.Y. Liang, A.R. Beal, A study of the optical joint density-of-states function, *Journal of Physics C: Solid State Physics* 9 (1976) 2823–2832. <https://doi.org/10.1088/0022-3719/9/14/020>.
- [132] M. Rashid, Study of Vanadium Difluoride VF_3 ($\text{A} = \text{Na, K, Rb}$) for Optoelectronic and Thermoelectric Device Applications via Ab Initio Calculations, *J Supercond Nov Magn* 33 (2020) 1167–1175. <https://doi.org/10.1007/s10948-019-05317-z>.
- [133] H. BENNACER, S. BERRAH, A. BOUKORTT, M.I. ZIANE, Electronic and optical properties of GaInX_2 ($\text{X} = \text{As, P}$) from first principles study, *Indian Journal*

- of Pure & Applied Physics (IJPAP) 53 (2015) 181–189.
<https://doi.org/10.56042/IJPAP.V53I3.5836>.
- [134] H.C. Kandpal, G.H. Fecher, C. Felser, Calculated electronic and magnetic properties of the half-metallic, transition metal based Heusler compounds, *J Phys D Appl Phys* 40 (2007) 1507. <https://doi.org/10.1088/0022-3727/40/6/S01>.
- [135] S.S. Oluyamo, D.B. Agunbiade, Spin Coated Tin Oxide Thin Film For Optical And Optoelectronic Material Applications, *International Journal of Innovated Research and Advanced Studies (IJIRAS)* 3 (2016) 394–398.
- [136] N.M. Saeed, Structural and optical properties of ZnS thin films prepared by spray pyrolysis technique, *Relation* 3 (2002) 2.
- [137] H. Lashgari, M.R. Abolhassani, A. Boochani, E. Sartipi, R. Taghavi-Mendi, A. Ghaderi, Ab initio study of electronic, magnetic, elastic and optical properties of full Heusler Co₂MnSb, *Indian Journal of Physics* 90 (2016) 909–916. <https://doi.org/10.1007/S12648-015-0829-Y/METRICS>.
- [138] A. Soussi, A. Ait Hssi, M. Boujnah, L. Boulkadat, K. Abouabassi, A. Asbayou, A. Elfanaoui, R. Markazi, A. Ihlal, K. Bouabid, Electronic and Optical Properties of TiO₂ Thin Films: Combined Experimental and Theoretical Study, *J Electron Mater* 50 (2021) 4497–4510. <https://doi.org/10.1007/s11664-021-08976-8>.
- [139] I.M. Baker, II-VI Narrow Bandgap Semiconductors: Optoelectronics, in: 2017: pp. 1–1. https://doi.org/10.1007/978-3-319-48933-9_34.
- [140] D. Comtesse, B. Geisler, P. Entel, P. Kratzer, L. Szunyogh, First-principles study of spin-dependent thermoelectric properties of half-metallic Heusler thin films between platinum leads, *Phys Rev B* 89 (2014) 094410. <https://doi.org/10.1103/PhysRevB.89.094410>.
- [141] S. Wimmer, D. Ködderitzsch, K. Chadova, H. Ebert, First-principles linear response description of the spin Nernst effect, *Phys Rev B* 88 (2013) 201108. <https://doi.org/10.1103/PhysRevB.88.201108>.
- [142] R. Freer, A. V. Powell, Realising the potential of thermoelectric technology: a Roadmap, *J Mater Chem C Mater* 8 (2020) 441–463. <https://doi.org/10.1039/C9TC05710B>.
- [143] J.R. Salvador, X. Shi, J. Yang, H. Wang, Synthesis and transport properties of M₃Ni₃Sb₄ (M=Zr and Hf): An intermetallic semiconductor, *Phys Rev B* 77 (2008) 235217. <https://doi.org/10.1103/PhysRevB.77.235217>.
- [144] Y.C. Gao, X. Gao, The half-metallicity of LiMgPdSn-type quaternary Heusler alloys FeMnScZ (Z=Al, Ga, In): A first-principle study, *AIP Adv* 5 (2015). <https://doi.org/10.1063/1.4921900/979972>.
- [145] M.D. Segall, P.J.D. Lindan, M.J. Probert, C.J. Pickard, P.J. Hasnip, S.J. Clark, M.C. Payne, First-principles simulation: ideas, illustrations and the CASTEP

- code, *Journal of Physics: Condensed Matter* 14 (2002) 2717–2744. <https://doi.org/10.1088/0953-8984/14/11/301>.
- [146] S.A. Khandy, J.-D. Chai, Novel half-metallic L21 structured full-Heusler compound for promising spintronic applications: A DFT-based computer simulation, *J Magn Magn Mater* 487 (2019) 165289. <https://doi.org/10.1016/j.jmmm.2019.165289>.
- [147] S.A. Khandy, Inspecting the electronic structure and thermoelectric power factor of novel p-type half-Heuslers, *Sci Rep* 11 (2021) 20756. <https://doi.org/10.1038/s41598-021-00314-6>.
- [148] S.A. Khandy, J.-D. Chai, Strain engineering of electronic structure, phonon, and thermoelectric properties of p-type half-Heusler semiconductor, *J Alloys Compd* 850 (2021) 156615. <https://doi.org/10.1016/j.jallcom.2020.156615>.
- [149] S.A. Khandy, J.-D. Chai, Origin of pseudo gap and thermoelectric signatures of semimetallic Ru₂TaGa: Structural stability from phonon dynamics, mechanical, and thermodynamic predictions, *Journal of Physics and Chemistry of Solids* 154 (2021) 110098. <https://doi.org/10.1016/j.jpcs.2021.110098>.
- [150] G.N. Greaves, A.L. Greer, R.S. Lakes, T. Rouxel, Poisson's ratio and modern materials, *Nat Mater* 10 (2011) 823–837. <https://doi.org/10.1038/nmat3134>.
- [151] I.N.; V.F.F.; B.S.A. Frantsevich, *Elastic Constants and Elastic Moduli of Metals and Insulators Handbook*, Naukova Dumka, Kyiv, Ukraine, 1983.
- [152] O.M. Abid, S. Menouer, A. Yakoubi, H. Khachai, S. Bin Omran, G. Murtaza, D. Prakash, R. Khenata, K.D. Verma, Structural, electronic, elastic, thermoelectric and thermodynamic properties of the NbMSb half heusler (M=Fe, Ru, Os) compounds with first principle calculations, *Superlattices Microstruct* 93 (2016) 171–185. <https://doi.org/10.1016/j.spmi.2016.01.001>.
- [153] A. Bakhshayeshi, M.M. Sarmazdeh, R.T. Mendi, A. Boochani, First-Principles Prediction of Electronic, Magnetic, and Optical Properties of Co₂MnAs Full-Heusler Half-Metallic Compound, *J Electron Mater* 46 (2017) 2196–2204. <https://doi.org/10.1007/S11664-016-5158-1/METRICS>.
- [154] M. Awais, F. Hussain, K.N. Riaz, N.A. Niaz, A. Shakoor, T. Dorigo, F. Ahmad, U. Mamtaz, First-principles calculations to investigate structural, electronic, and optical properties of AgWO₄ and PdWO₄ tungstate materials for optoelectronic applications, *Next Materials* 4 (2024) 100113. <https://doi.org/10.1016/j.nxmte.2024.100113>.
- [155] T. Stauber, N.M.R. Peres, A.K. Geim, Optical conductivity of graphene in the visible region of the spectrum, *Phys Rev B* 78 (2008) 085432. <https://doi.org/10.1103/PhysRevB.78.085432>.
- [156] M. Roknuzzaman, K. (Ken) Ostrikov, K. Chandula Wasalathilake, C. Yan, H. Wang, T. Tesfamichael, Insight into lead-free organic-inorganic hybrid

- perovskites for photovoltaics and optoelectronics: A first-principles study, *Org Electron* 59 (2018) 99–106. <https://doi.org/10.1016/j.orgel.2018.04.051>.
- [157] Marti'nez-Duart, R.J. Marti'nez-Palma, Photodetectors and Solar Cells Based on Porous Silicon, *Physica Status Solidi (b)* 232 (2002) 81–88. [https://doi.org/https://doi.org/10.1002/1521-3951\(200207\)232:1<81::AID-PSSB81>3.0.CO;2-E](https://doi.org/https://doi.org/10.1002/1521-3951(200207)232:1<81::AID-PSSB81>3.0.CO;2-E).
- [158] E. Haque, M.A. Hossain, Electronic, phonon transport and thermoelectric properties of Cs₂InAgCl₆ from first-principles study, *Computational Condensed Matter* 19 (2019) e00374. <https://doi.org/10.1016/j.cocom.2019.e00374>.
- [159] G. Kresse, J. Furthmüller, Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set, *Phys Rev B* 54 (1996) 11169–11186. <https://doi.org/10.1103/PhysRevB.54.11169>.
- [160] S. Singh, S. Singh, N. Bijewar, A. Kumar, Thermoelectric properties of half-metallic FeMnScGa using first principle calculation, in: *AIP Conf Proc*, 2020.
- [161] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G.L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A.P. Seitsonen, A. Smogunov, P. Umari, R.M. Wentzcovitch, QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials, *Journal of Physics: Condensed Matter* 21 (2009) 395502. <https://doi.org/10.1088/0953-8984/21/39/395502>.

Appendices

Appendix A

Figures

Figure A.1

(a-d) Dielectric function of FeMnCrGe using mBJ and GGA methods

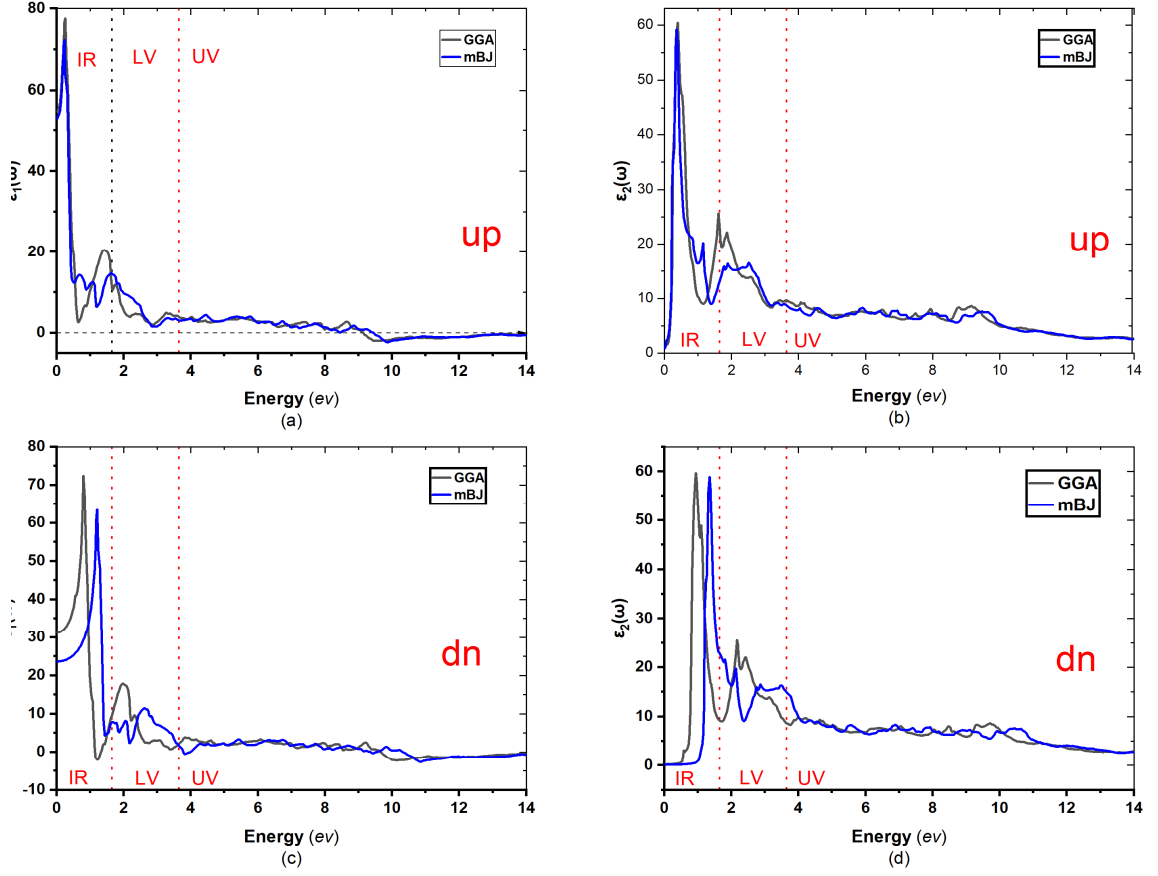


Figure A.2

Spin-dependent analysis of refractive index and extinction coefficient with GGA and mBJ.

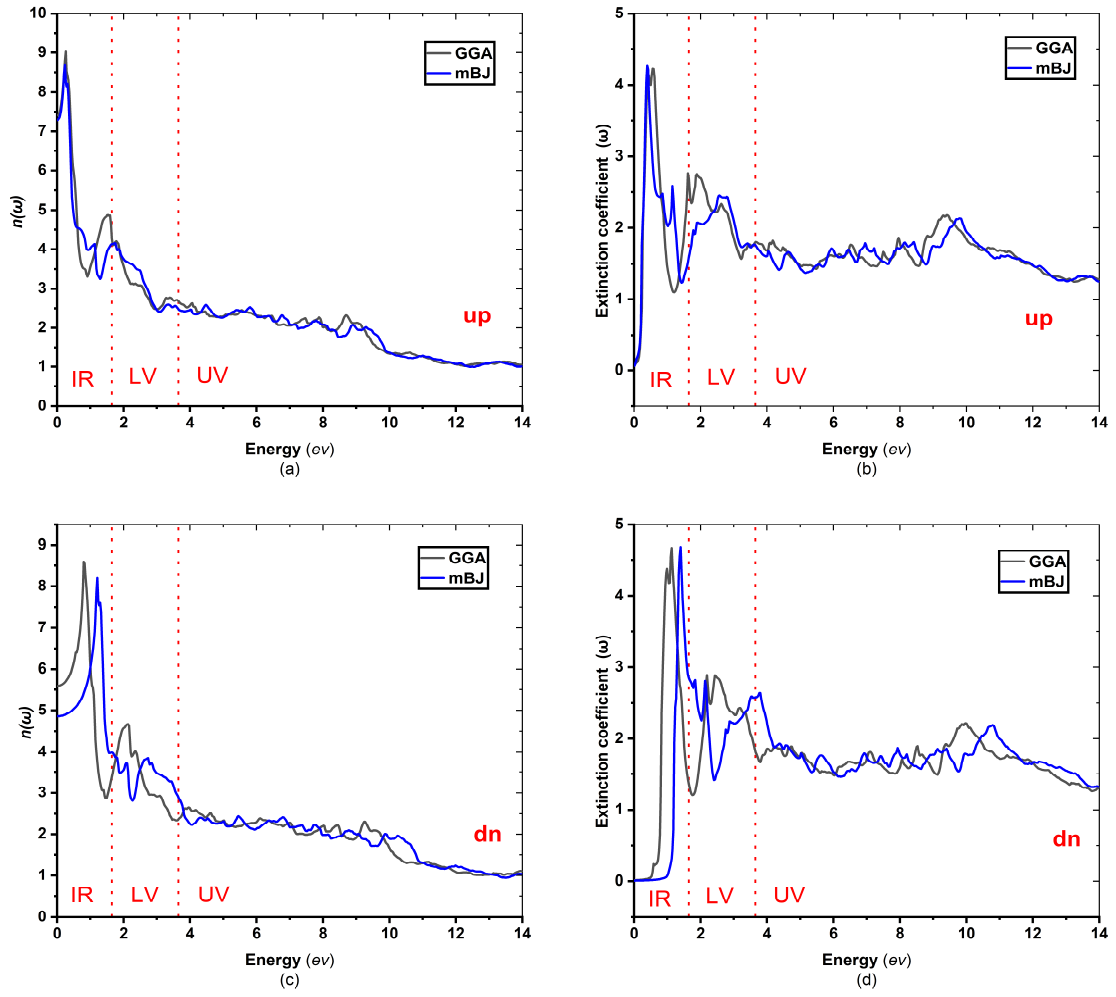


Figure A.3

Absorption coefficient of FeMnCrGe across spin channels using GGA and mBJ approximations

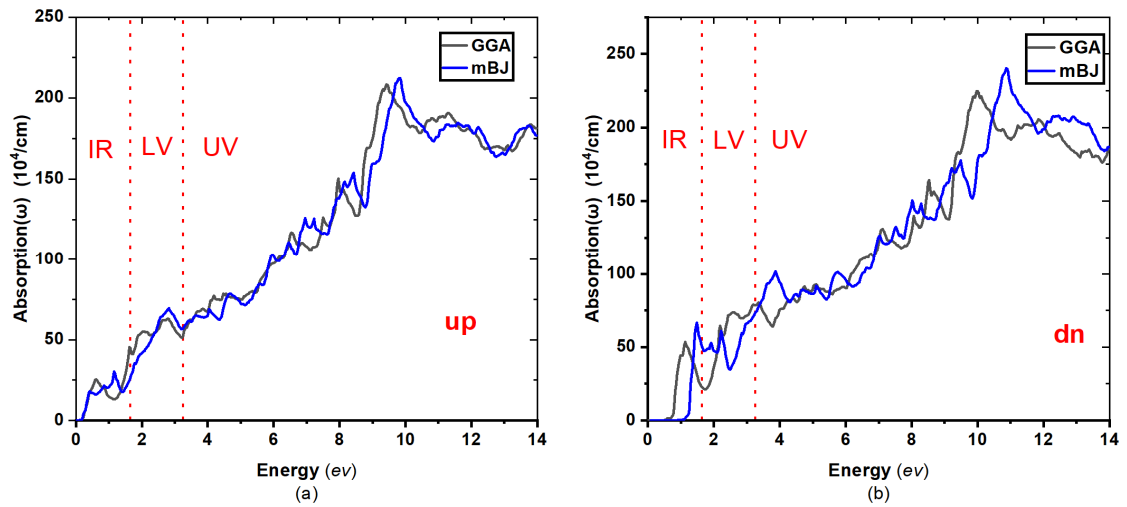


Figure A.4

Optical conductivity of FeMnCrGe in spin configurations using GGA and mBJ

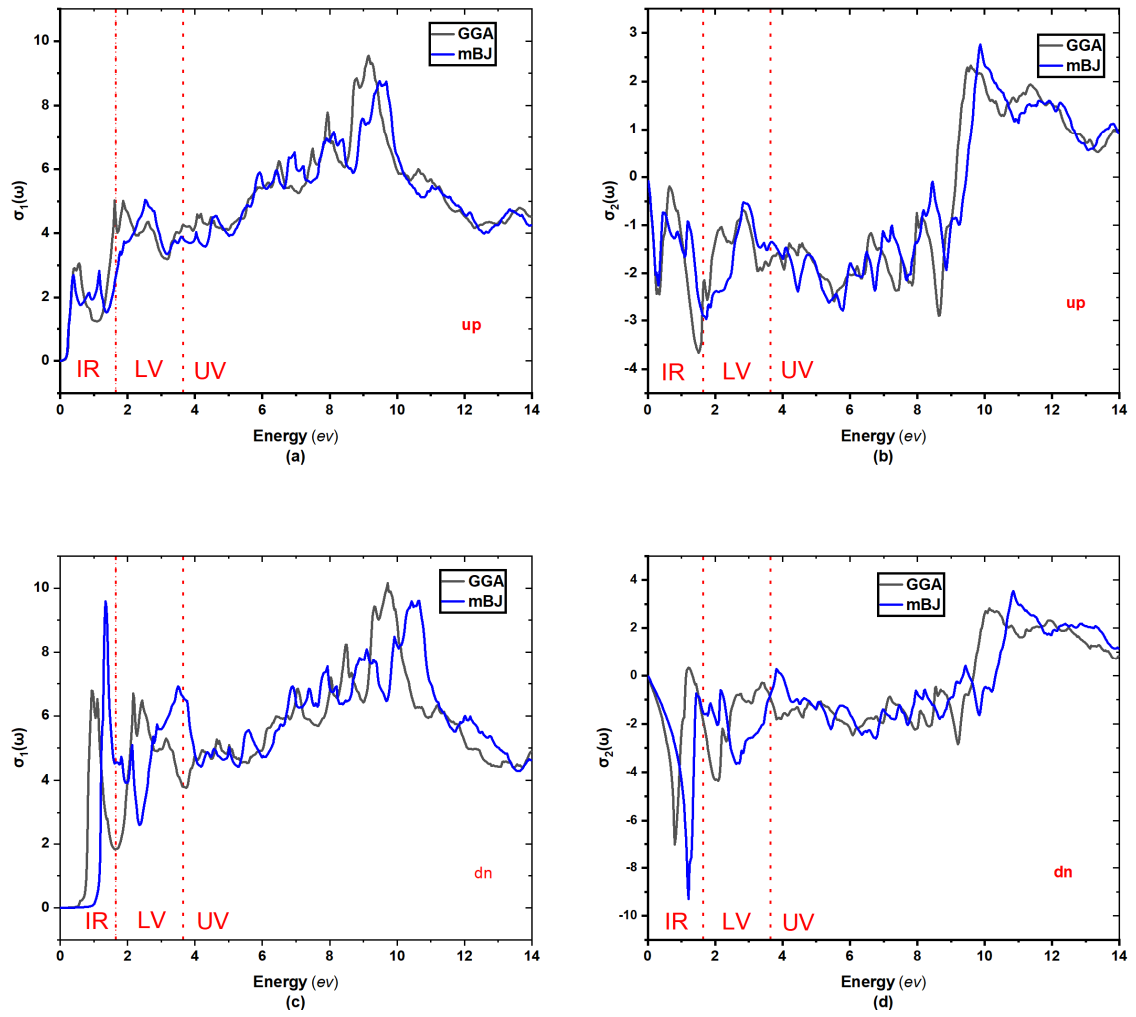


Figure A.5

Reflectivity spectrum of FeMnCrGe for Spin channels using GGA and mBJ Methods

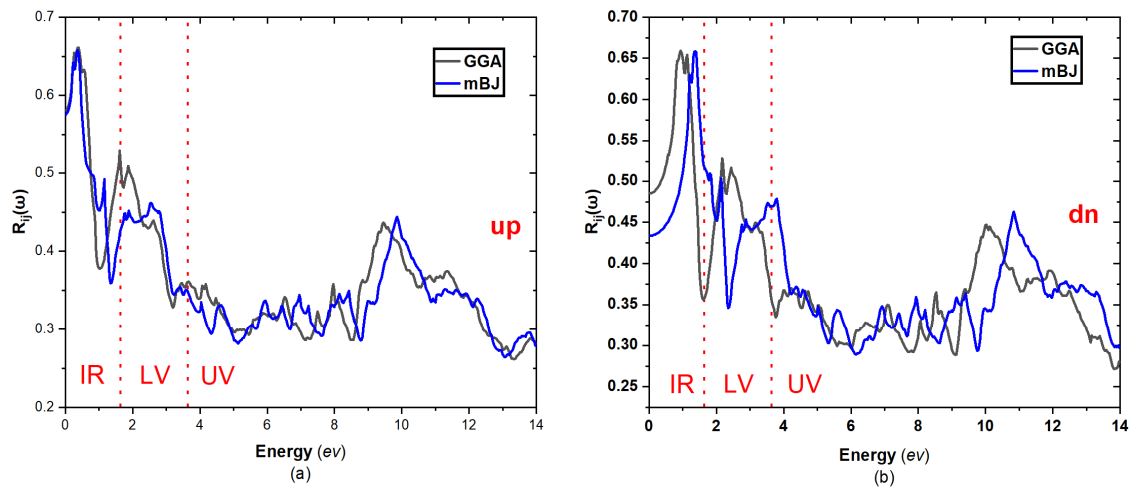


Figure A.6

Energy Loss function of FeMnCrGe in spin configurations using GGA and mBJ

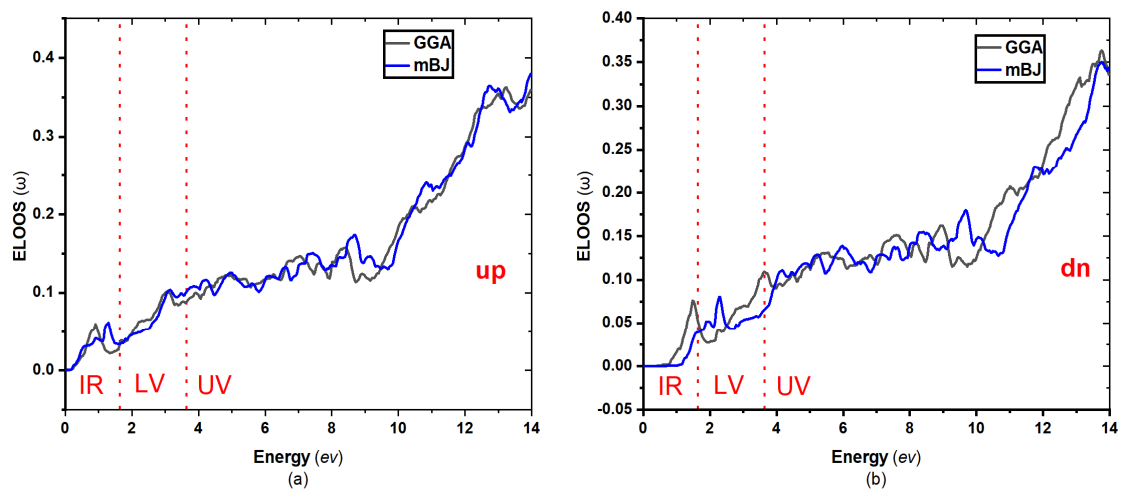


Figure A.7

(a-h) Temperature-dependent thermoelectric properties of FeMnCrGe for spin-up and spin-down channels (GGA vs. mBJ)

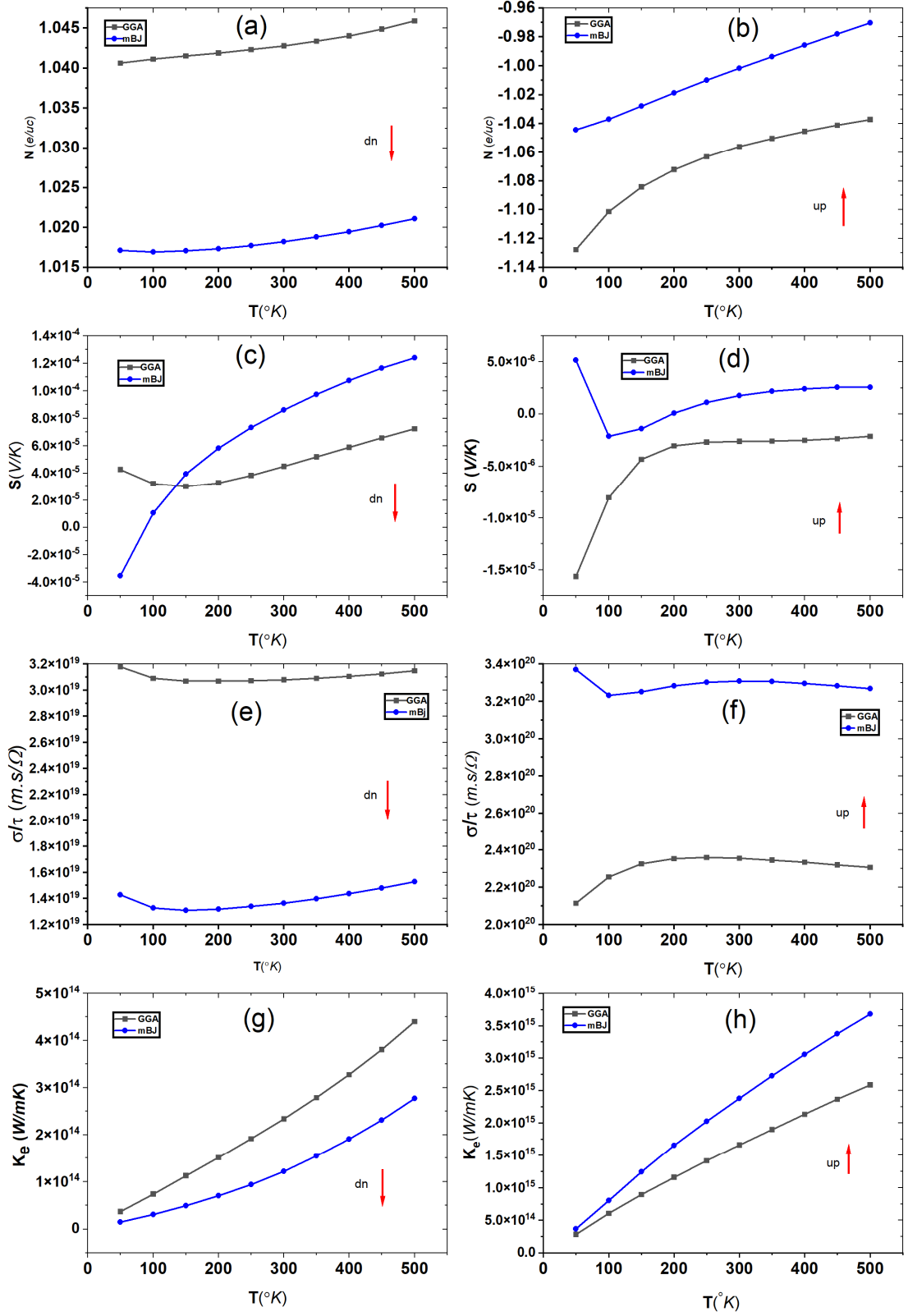


Figure A.8

(a) & (b) Thermoelectric Figure of Merit (ZT) as a function of temperature for FeMnCrGe (GGA vs. mBJ Methods)

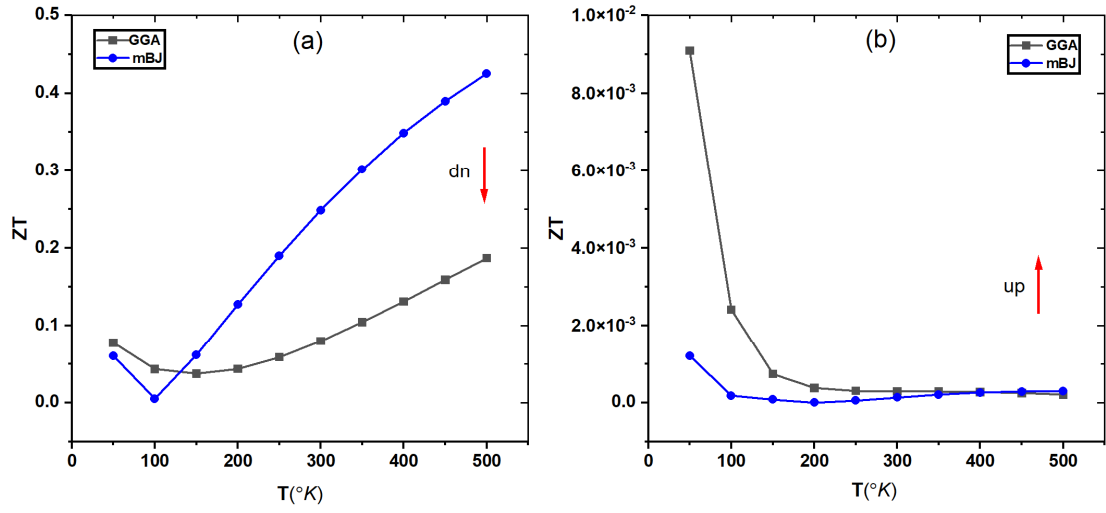


Figure A.9

(a) & (b) Temperature dependence of Power Factor (PF) for FeMnCrGe (GGA vs. mBJ Methods)

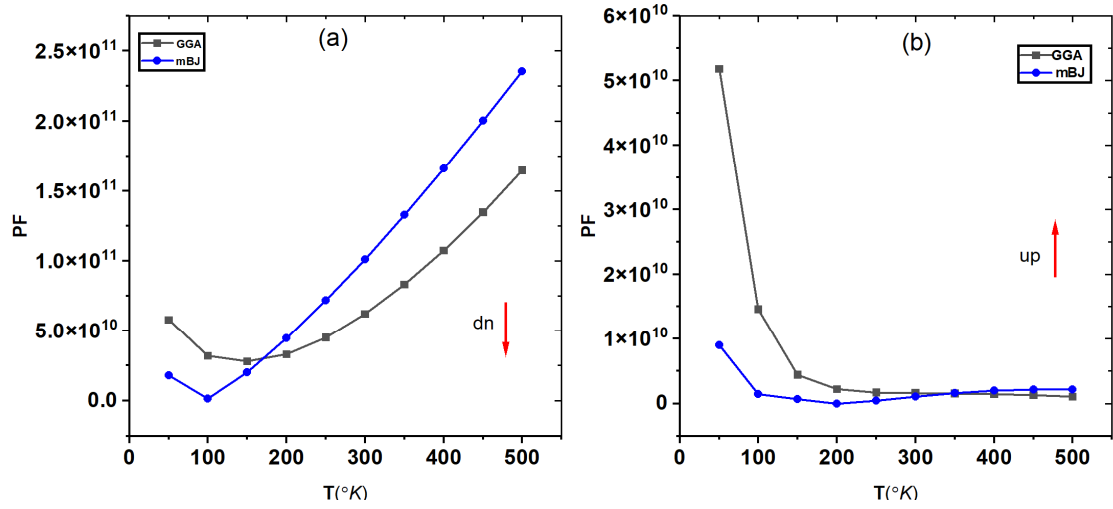


Figure A.10

(a-b) Hall Coefficient (R_H) variation with temperature in FeMnCrGe (GGA vs. mBJ Methods)

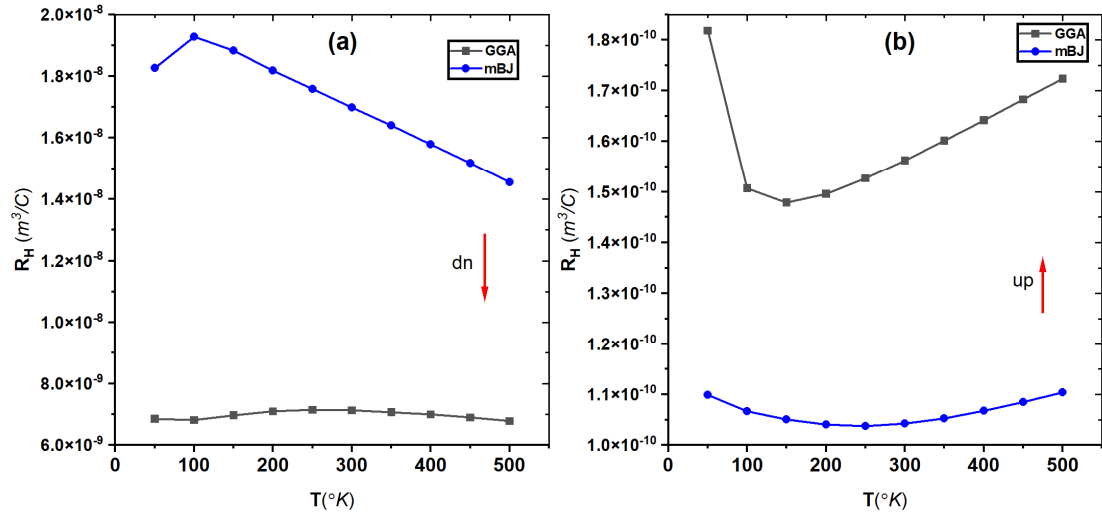


Figure A.11

Temperature-Dependent Variation of Lattice Thermal Conductivity in FeMnCrGe Based on the Slack Model.

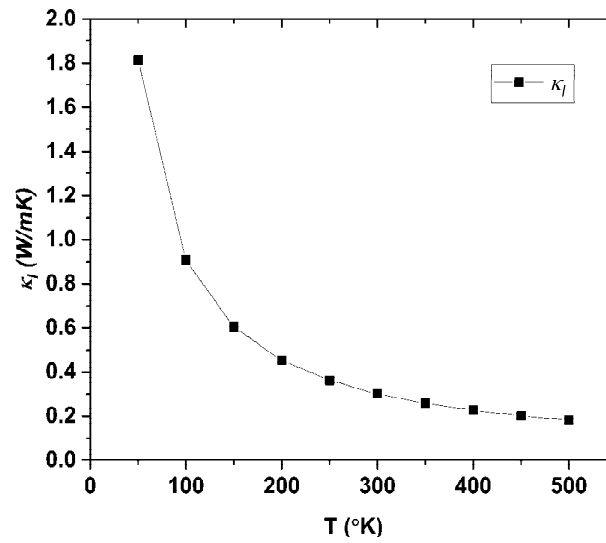


Figure A.12

Temperature Dependence of Total Thermal Conductivity in FeMnCrGe for Majority and Minority Spin Channels

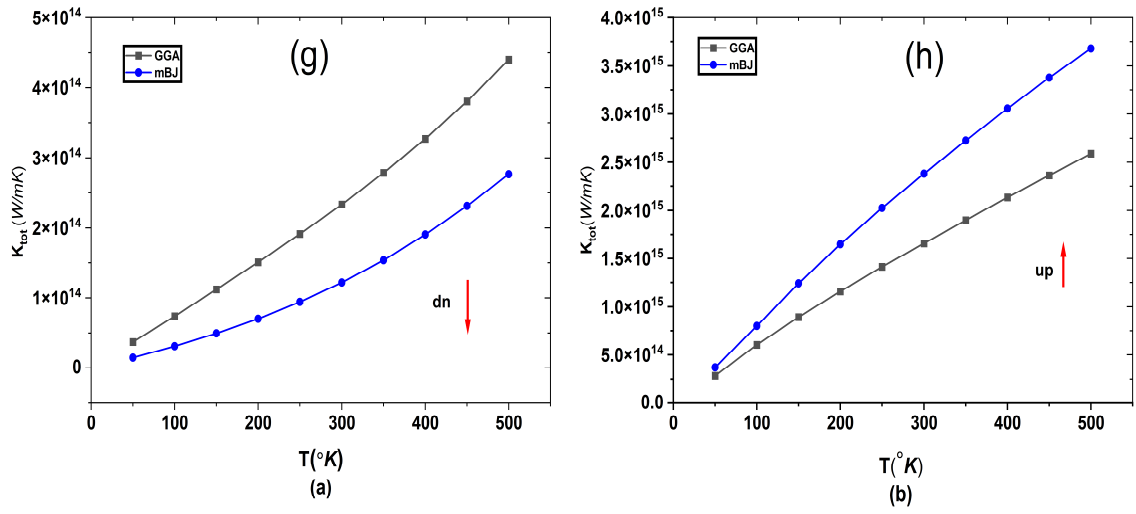


Figure A.13

The total energy as a function of unit cell volume for three FeMnScAl configurations is presented

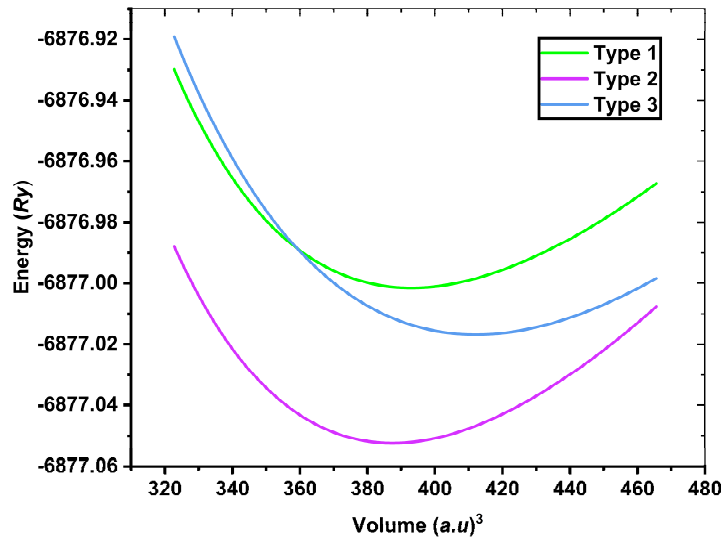


Figure A.14

The three FeMnScAl crystal structures along with their Brillouin zone and high-symmetry points

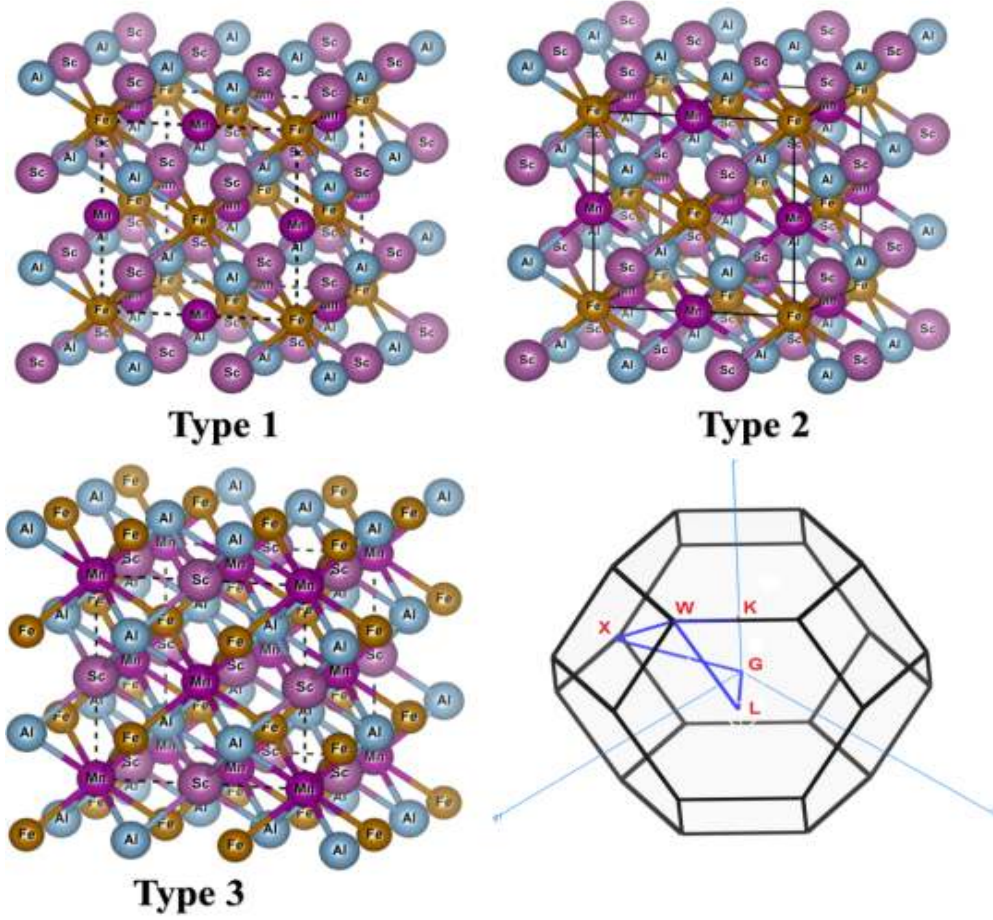


Figure A.15

The phonon dispersion curve for FeMnScAl along the high-symmetry paths in the Brillouin zone

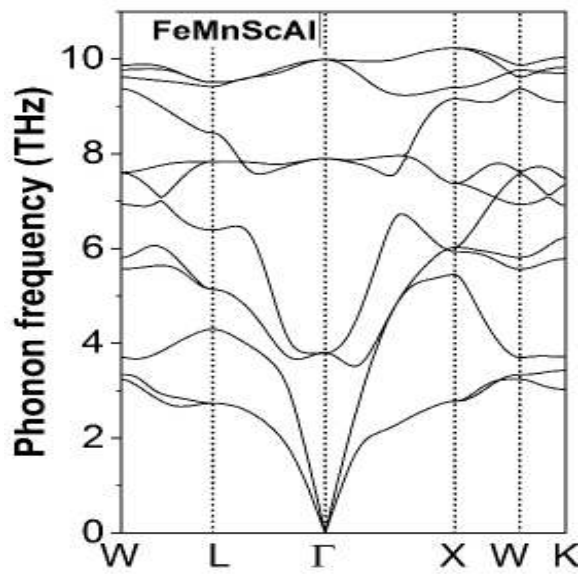


Figure A.16

(a) and (b) Contour plots of charge density for the FeMnScAl alloy

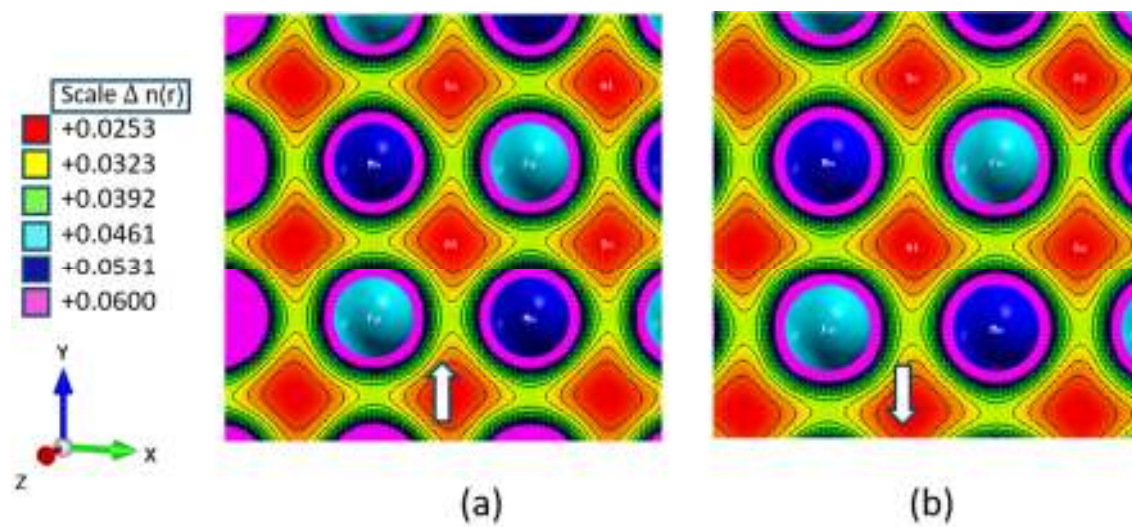


Figure A.17

(a-e) Density of states (DOS) of FeMnScAl with total and atomic contributions, computed using the GGA approximation

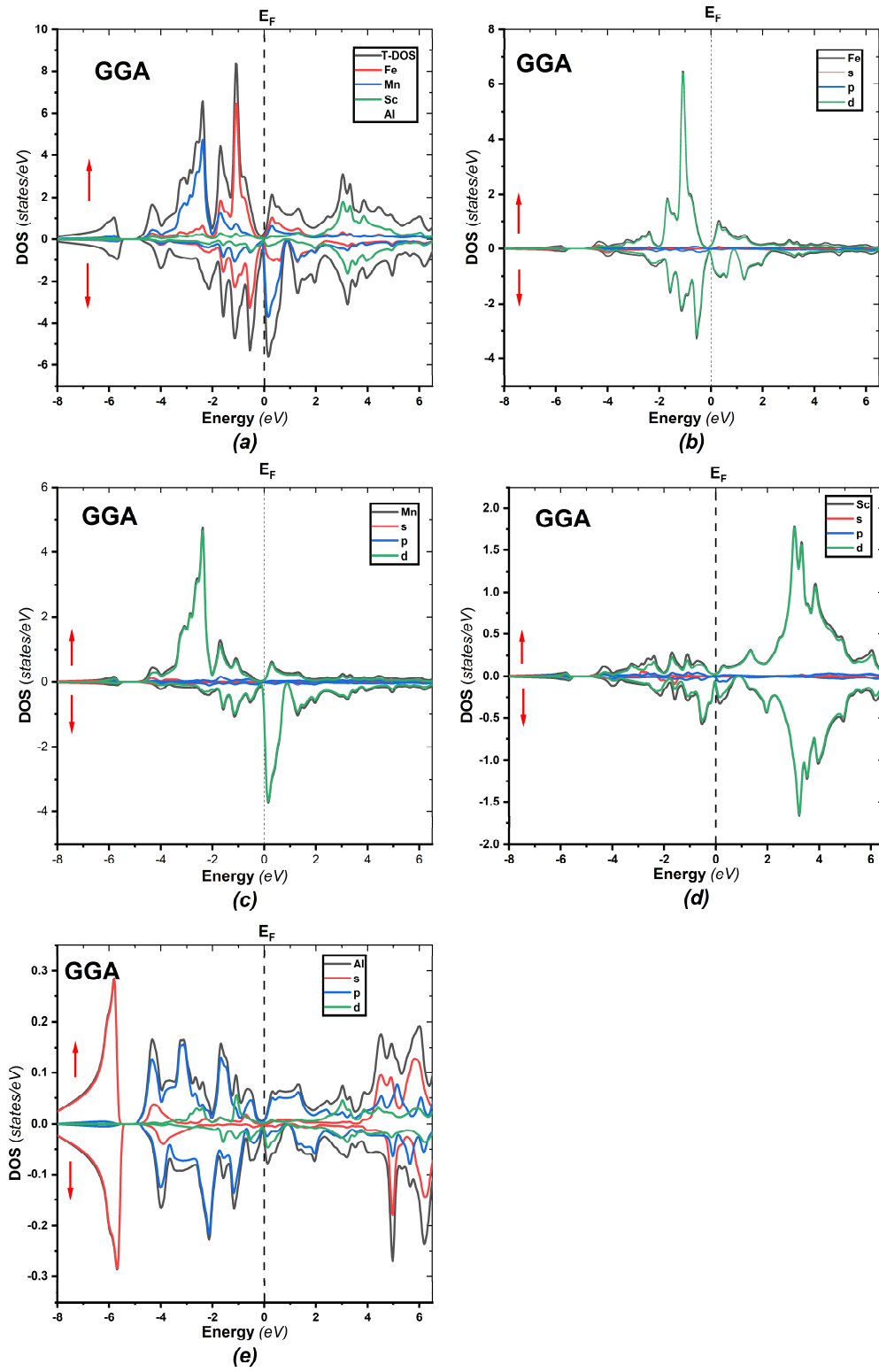


Figure A.18

(a-e) Density of states (DOS) of FeMnScAl, including total and atomic contributions, computed with the mBJ approximation

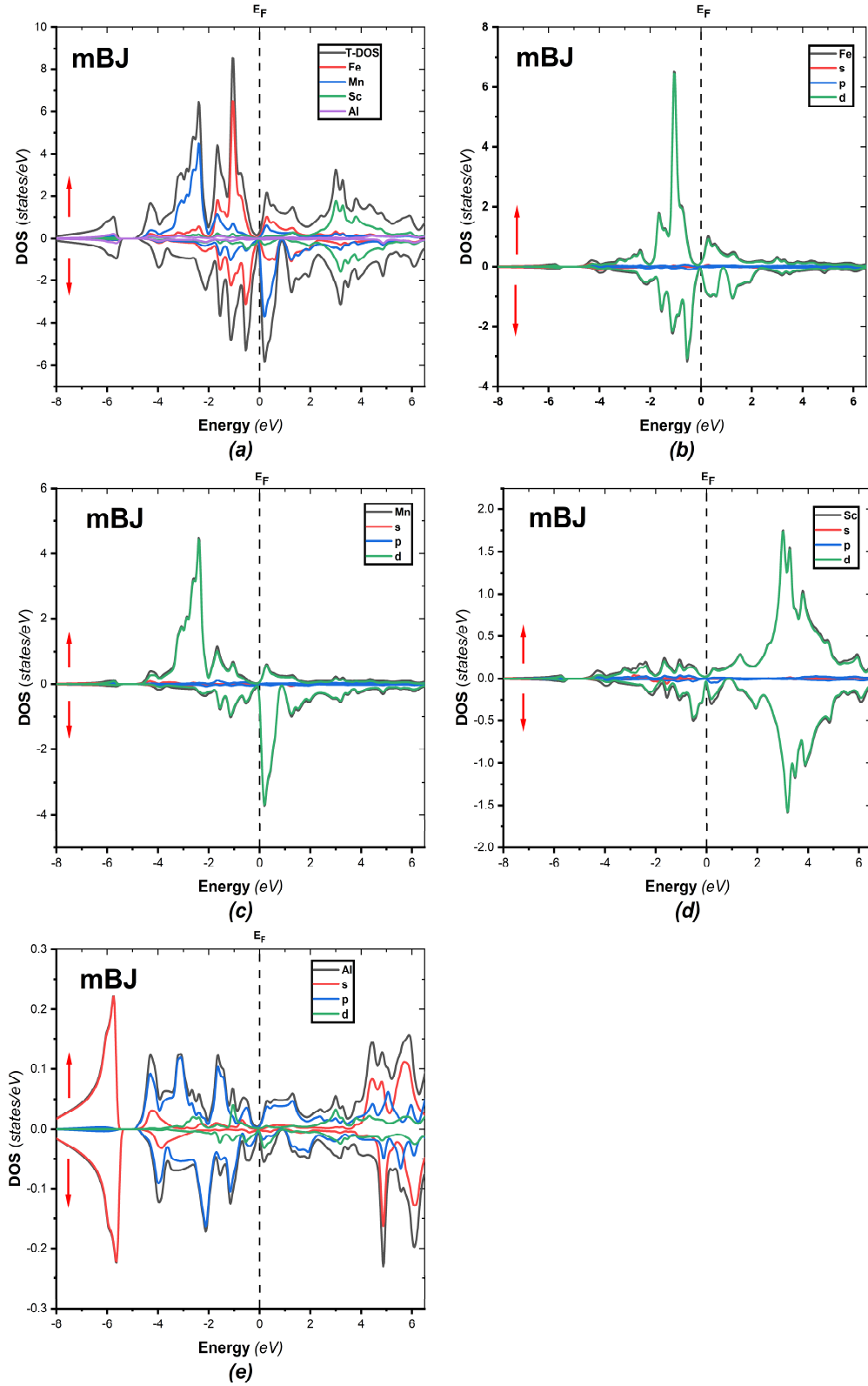


Figure A.19

(a-d) Electronic band structures of FeMnScAl with spin polarization, computed using GGA and mBJ

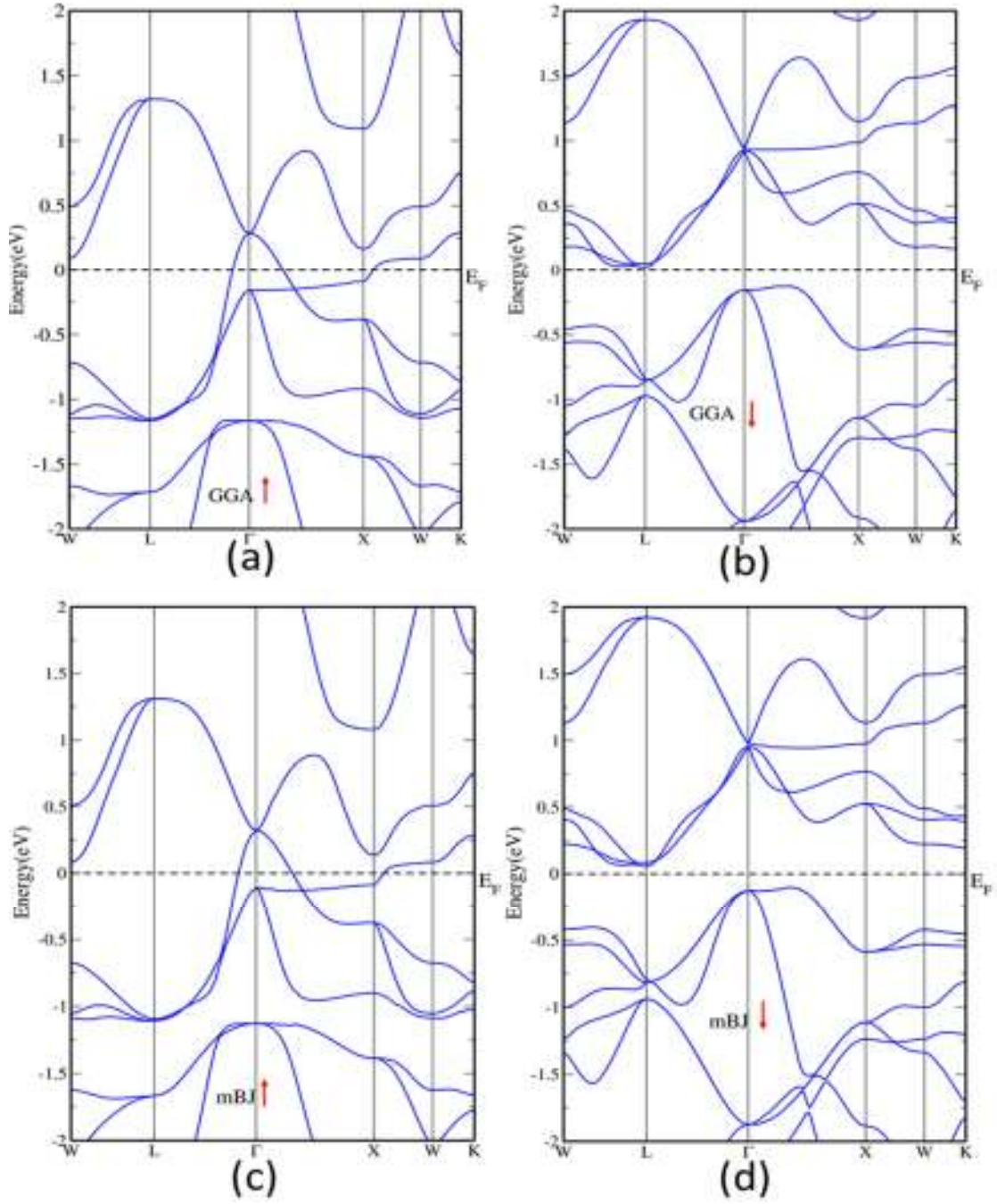


Figure A.20

Energy vs. Magnetic Configuration for FeMnScAl (AFM, FM, PM)

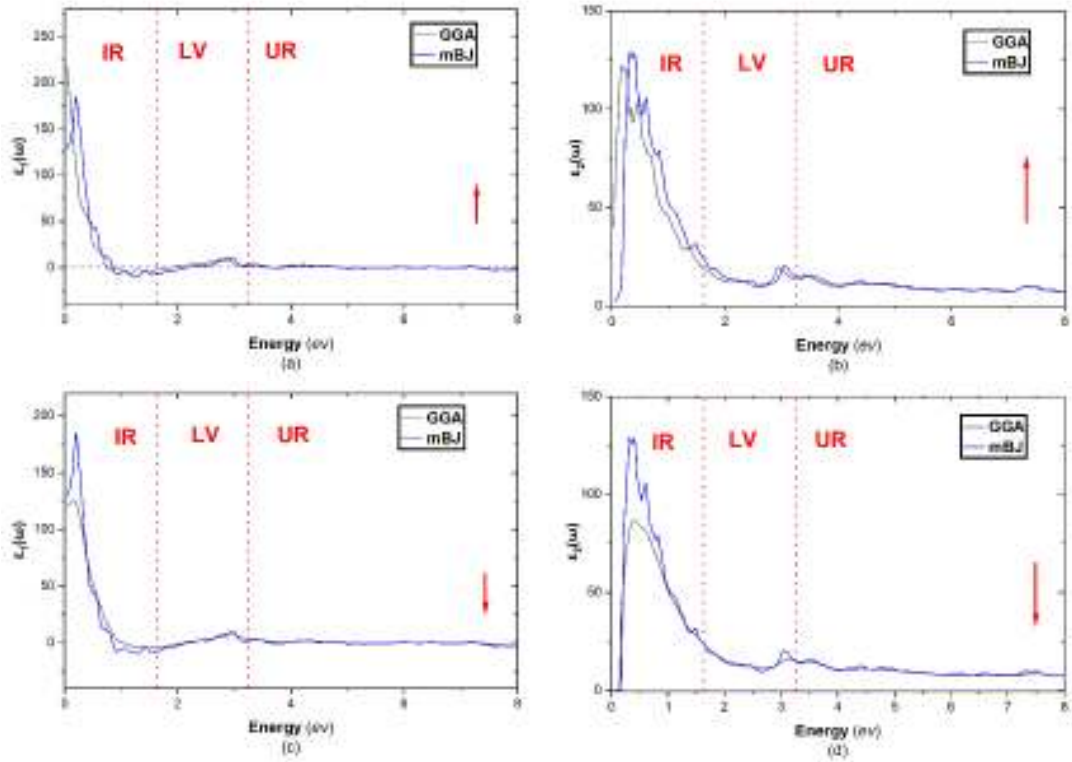


Figure A.21

(a) and (b) FeMnScAl's dielectric function determined using GGA and mBJ methods.

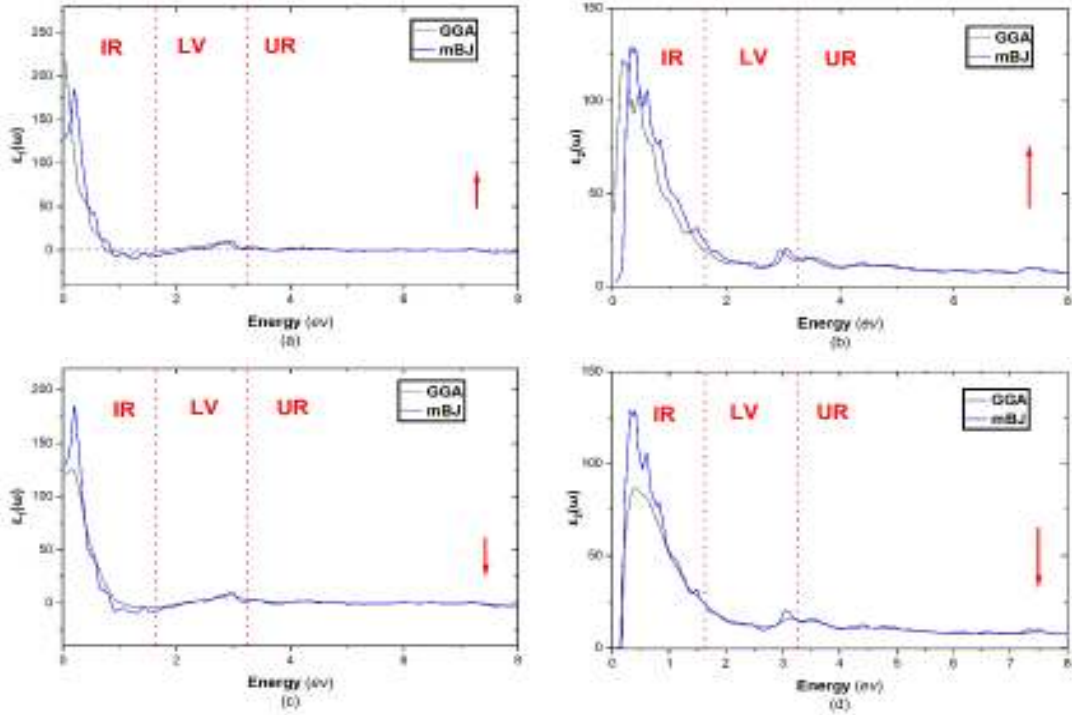


Figure A.22

(a) The refractive index as a function of energy and (b) the extinction coefficient of FeMnScAl calculated using mBJ and GGA methods.

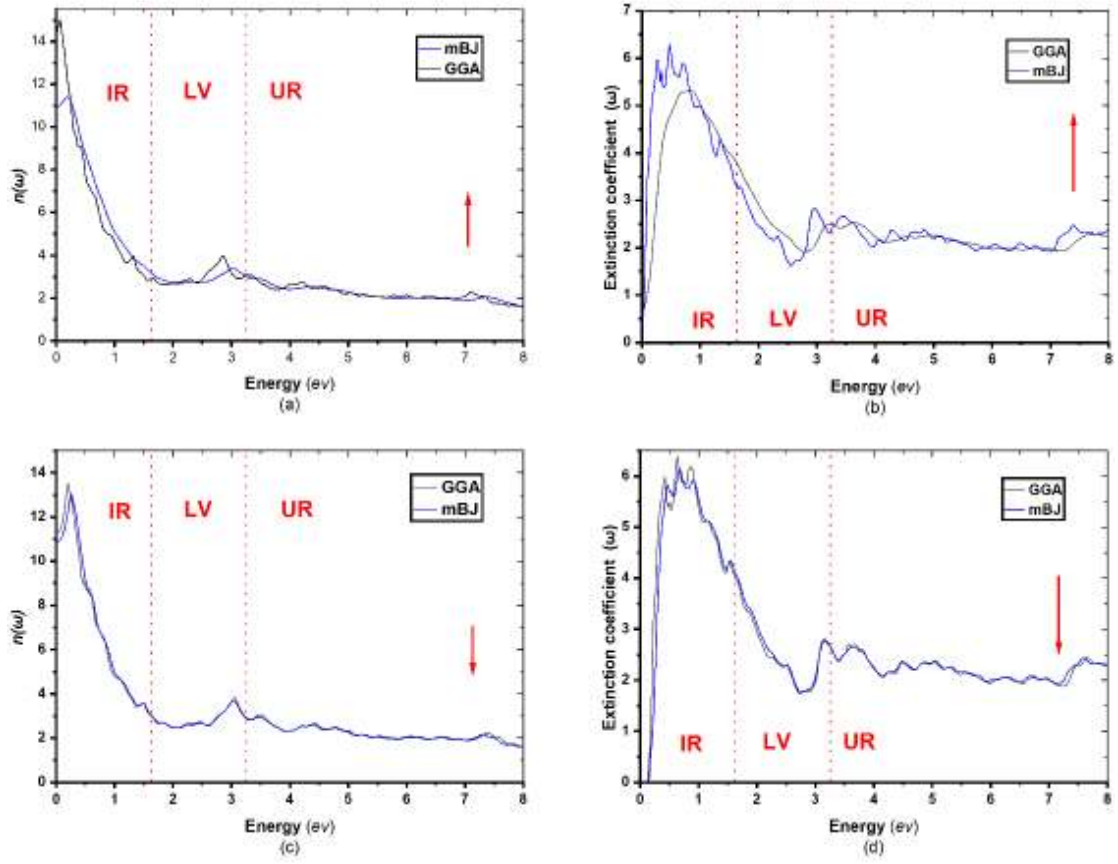


Figure A.23

The spectrum of absorption coefficient $A(\omega)$ for FeMnScAl alloy, calculated with mBJ and GGA methods

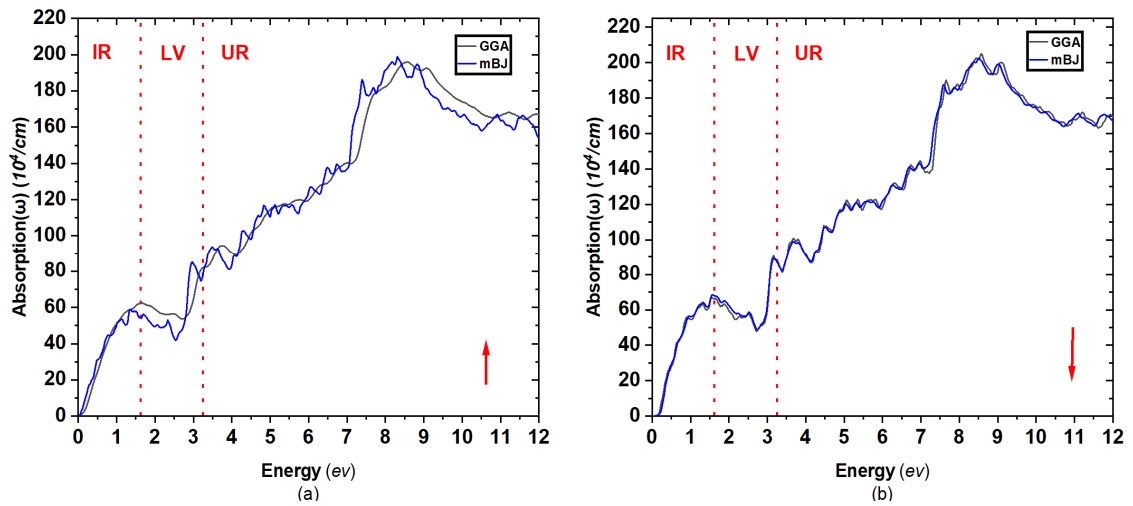


Figure A.24

(a) and (b) The FeMnScAl alloy's $\sigma(\omega)$ optical conductivity spectrum, assessed using mBJ and GGA techniques

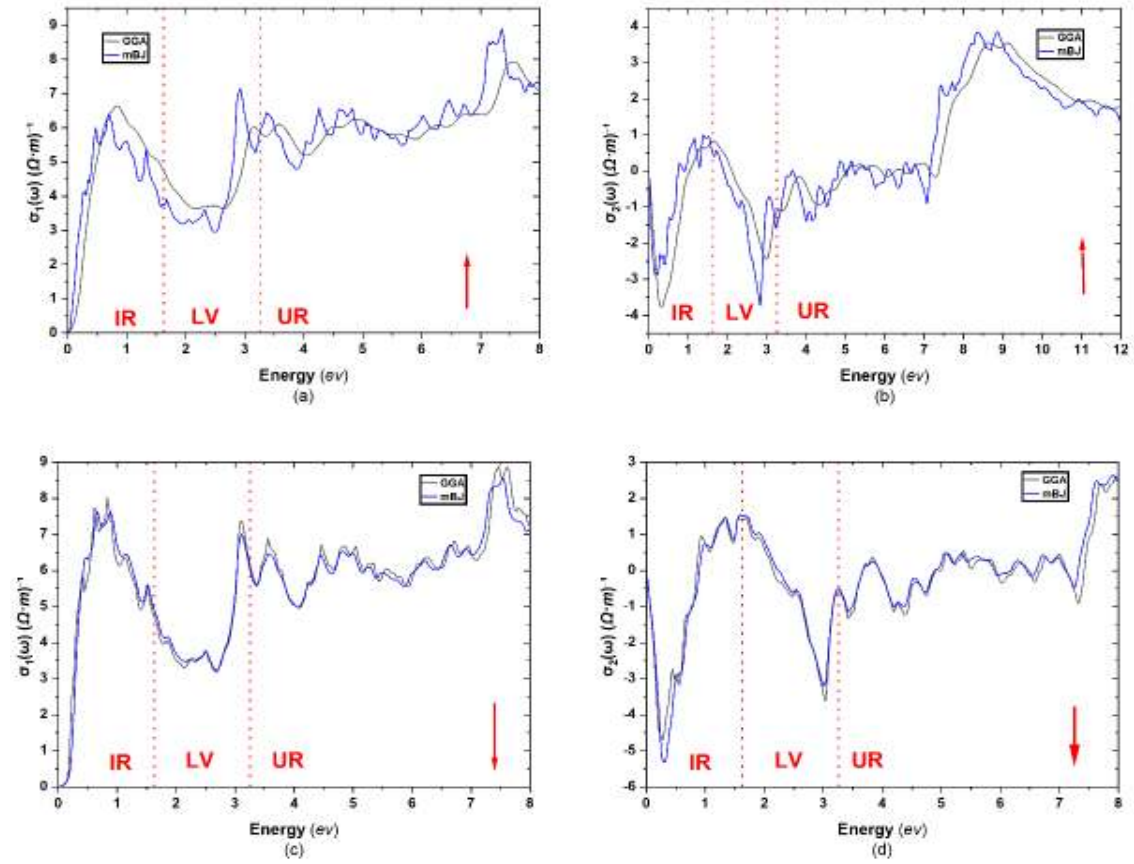


Figure A.25

The reflectivity spectrum of FeMnScAl for different spin channels, obtained through GGA and mBJ approaches

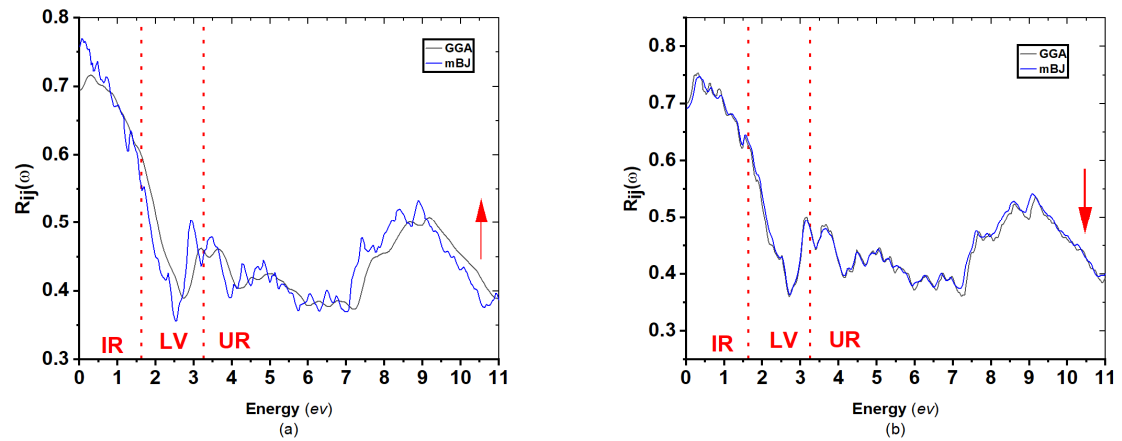


Figure A.26

The energy loss function $L(\omega)$ spectrum of FeMnScAl alloy, studied with mBJ and GGA approaches

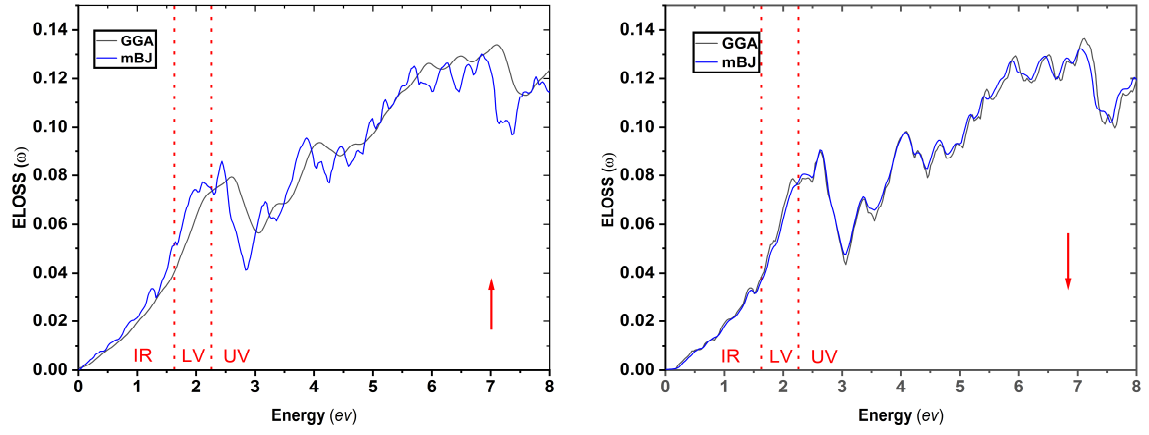


Figure A.27

(a-h) Thermoelectric Characteristics for Spin-Up and Spin-Down Channels of FeMnScAl Alloy (GGA and mBJ)

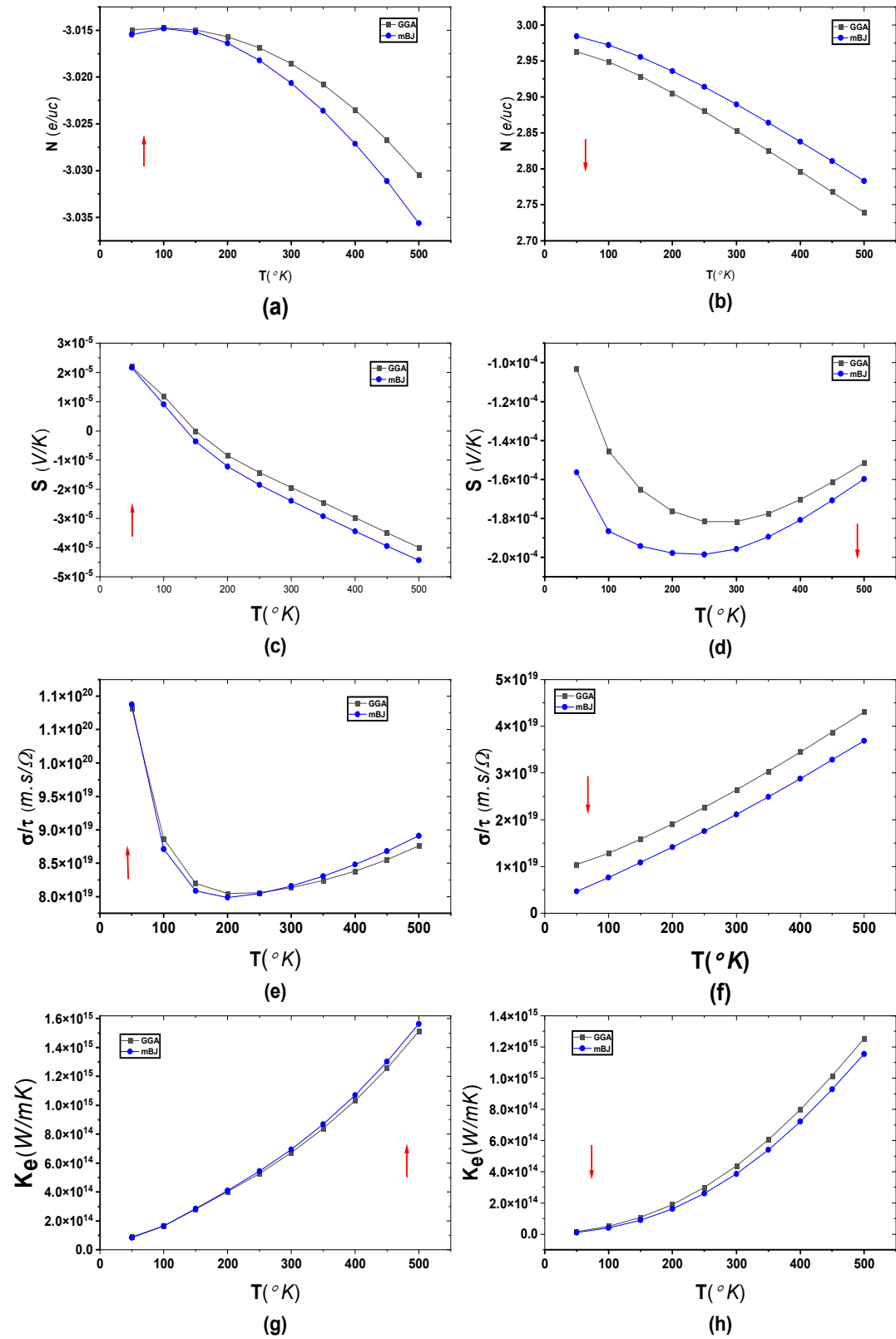


Figure A.28

Thermoelectric performance metrics (ZT and PF) for spin-up and spin-down channels (GGA and mBJ)

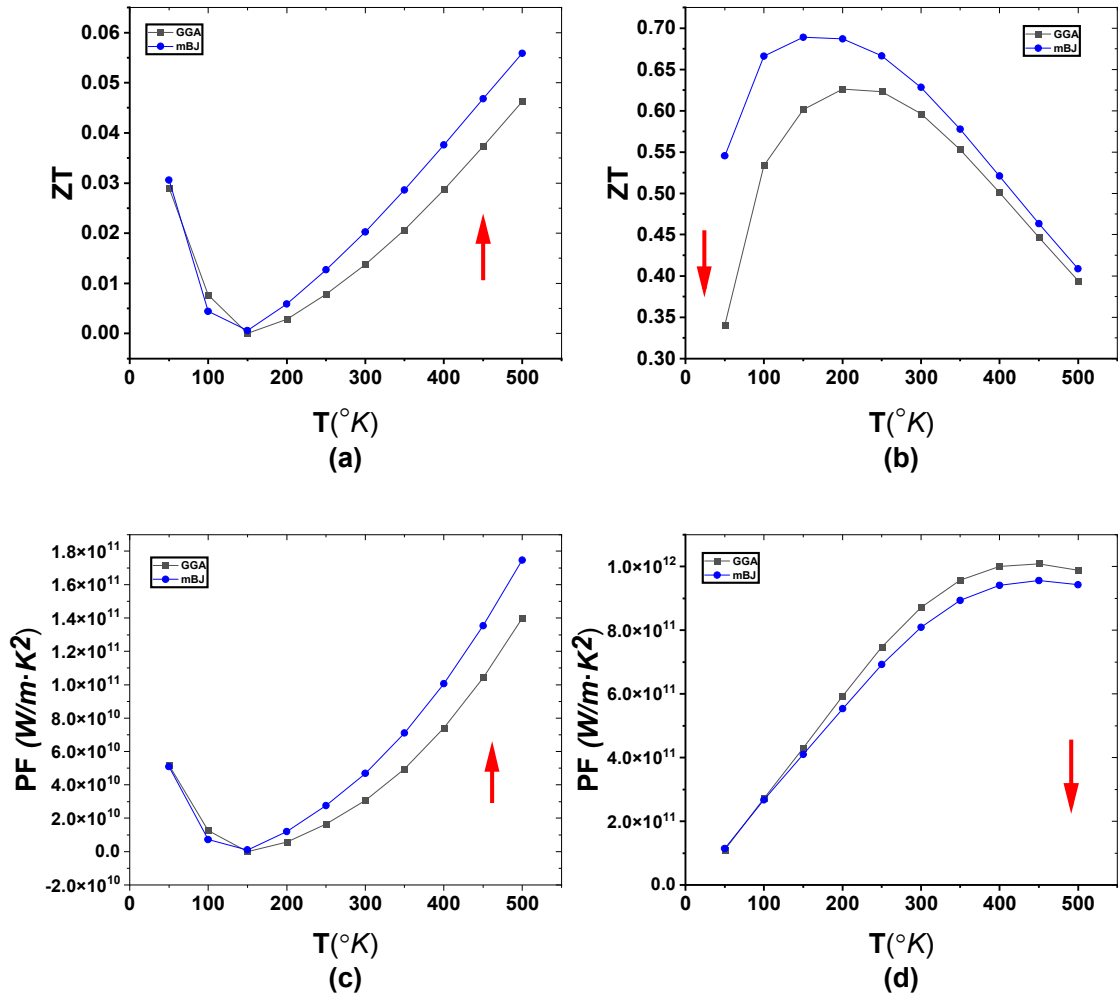


Figure A.29

Hall Coefficient (R_H) of FeMnScAl Alloy for spin-up and Spin-down channels (GGA and mBJ)

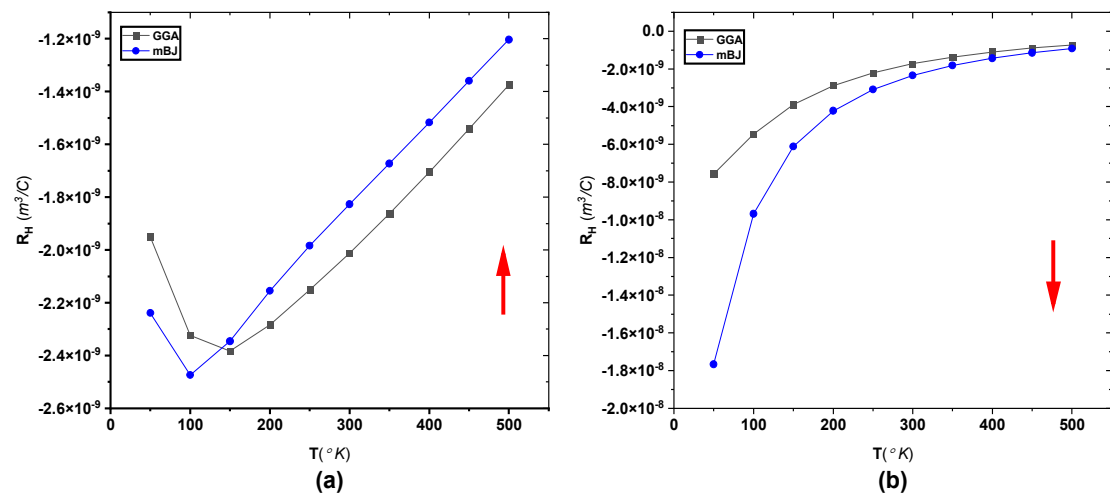


Figure A.30

Lattice and total thermal conductivity (κ) of FeMnScAl alloy at varying Temperatures (GGA and mBJ)

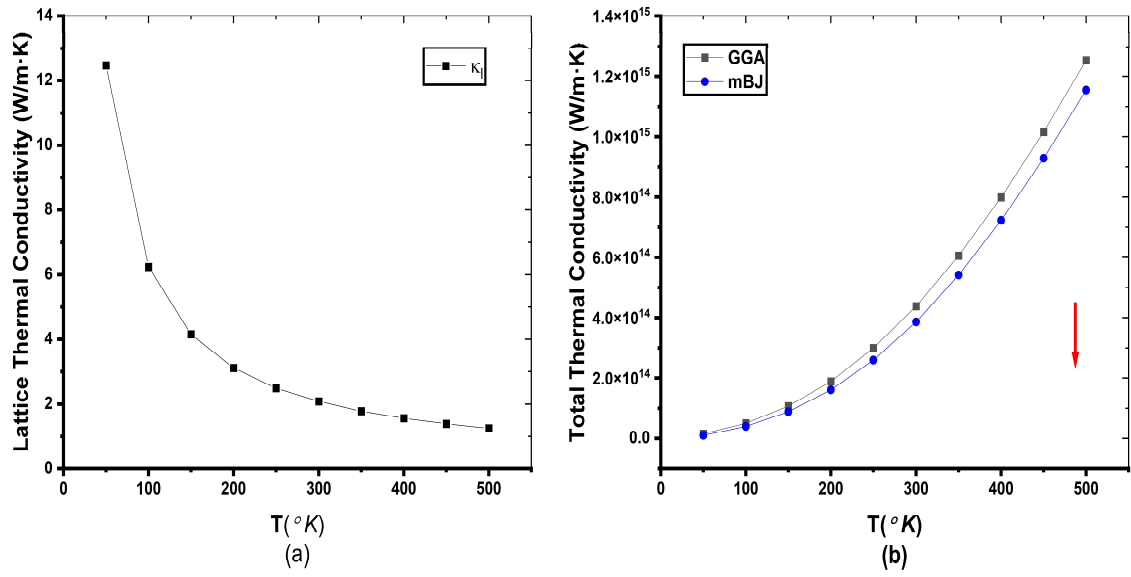


Figure A.31

Three cubic structural configurations, labeled Types 1, 2, and 3, represent the atomic arrangements of FeMnScGa.

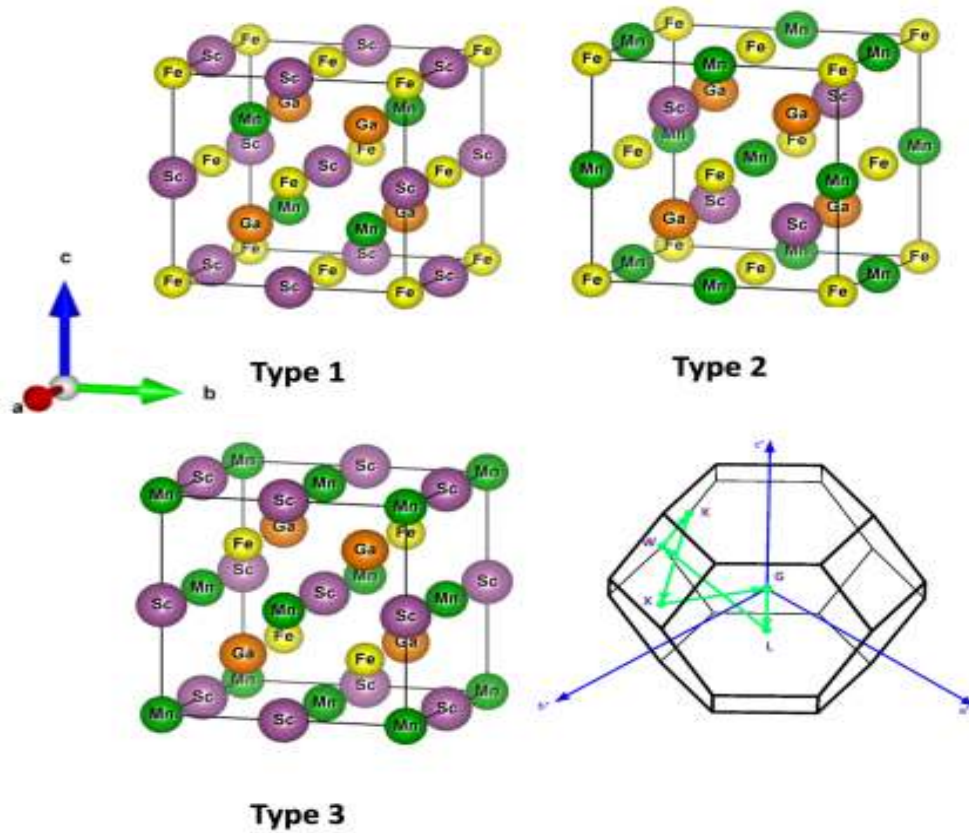


Figure A.32

Total energy variation with unit cell volume for FeMnScGa types

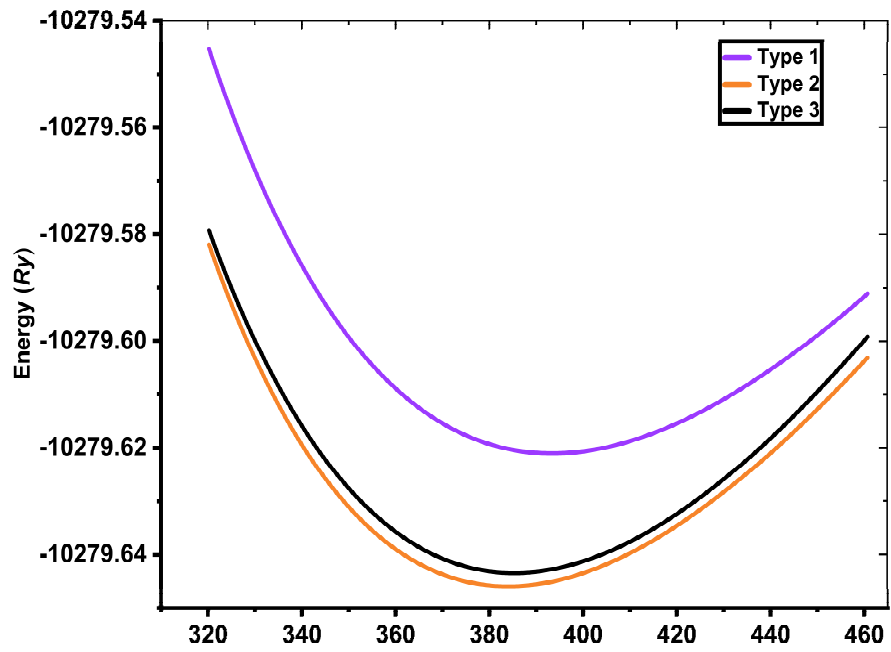


Figure A.33

Phonon Spectral Distribution of FeMnScGa

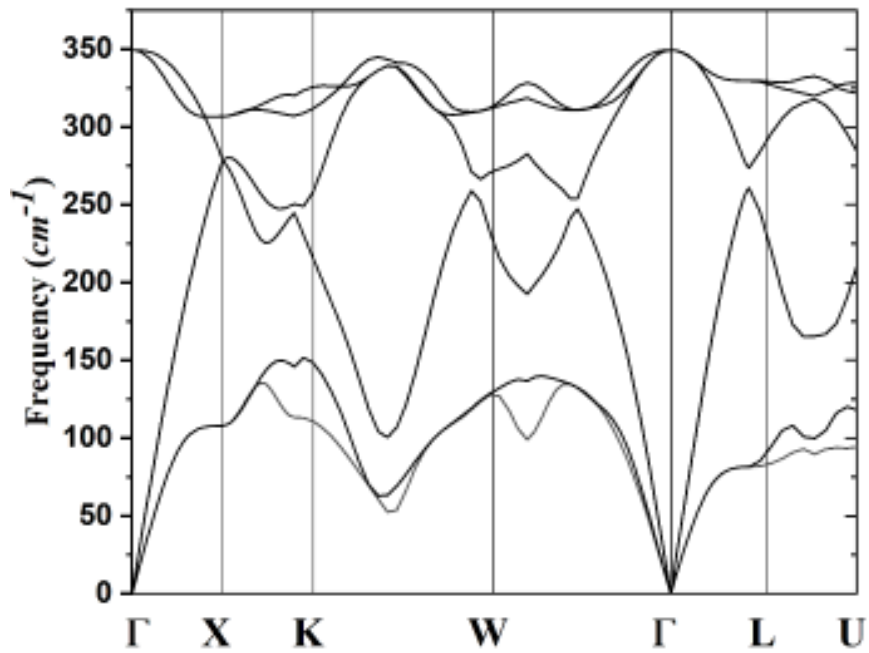


Figure A.34

(a-e) GGA-Based Analysis of Total and Atomic DOS Contributions in FeMnScGa

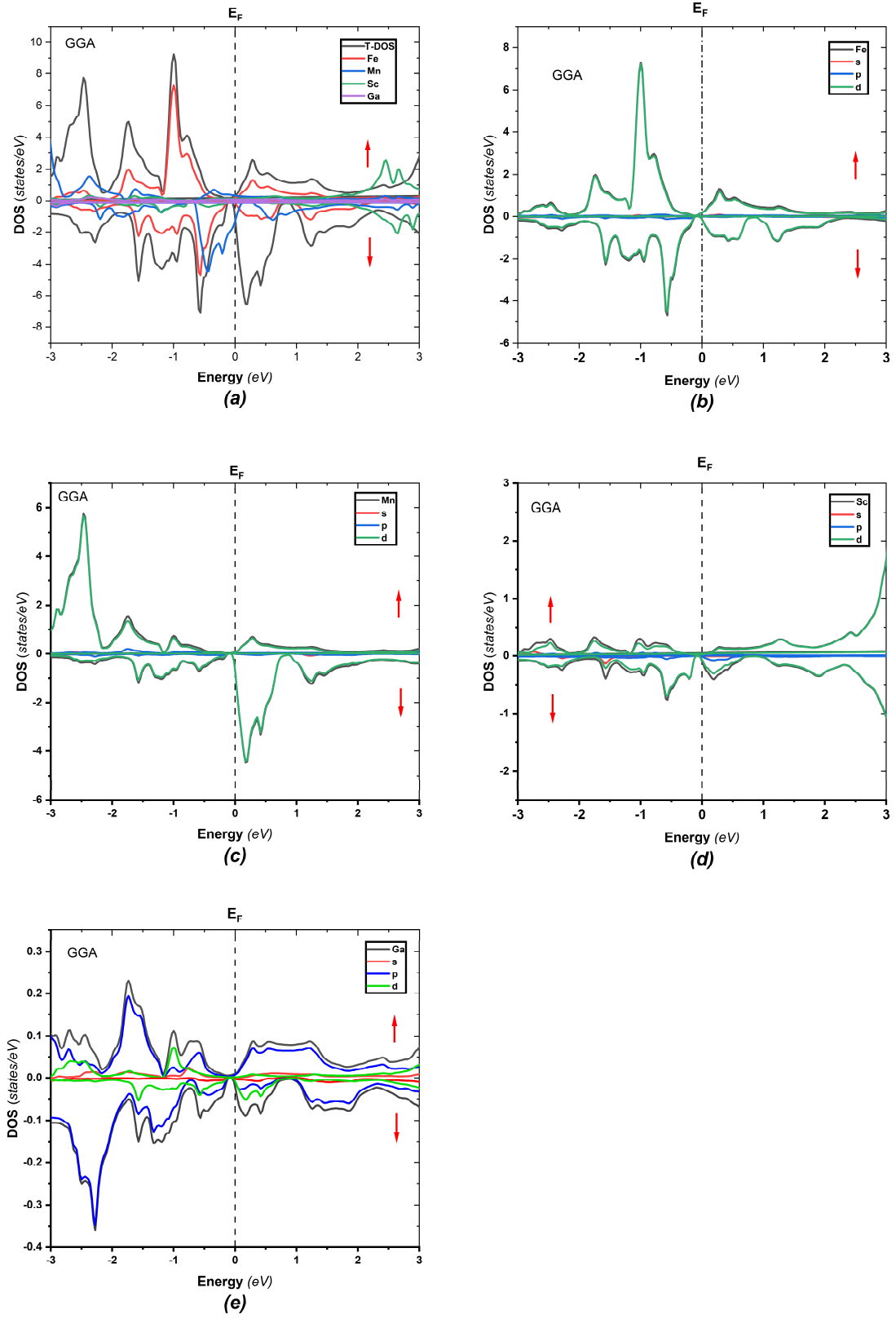


Figure A.35

(a-e) mBJ-Based Analysis of Total and Atomic DOS Contributions in FeMnScGa

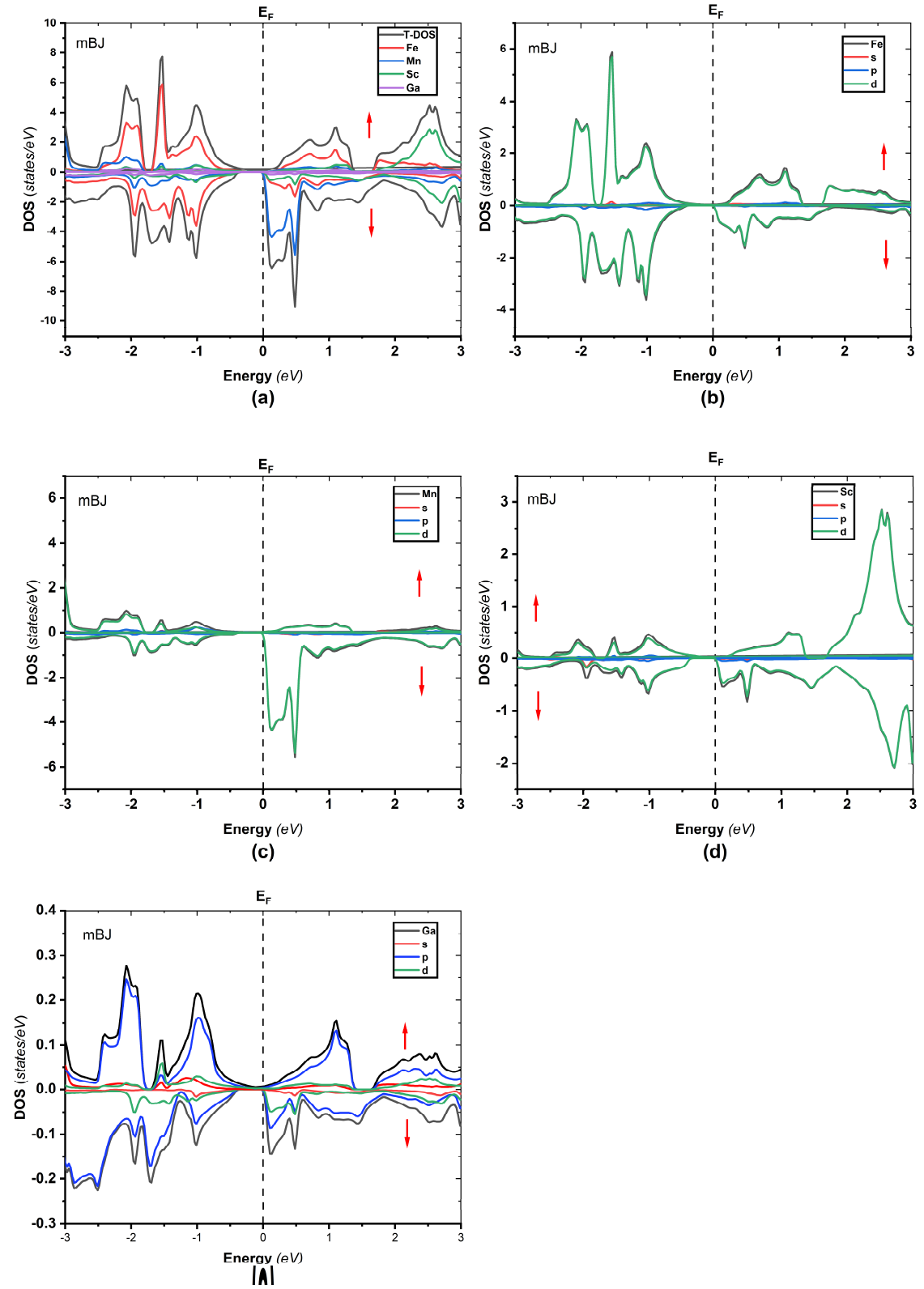


Figure A.36

Spin-polarized electronic band structures of FeMnScGa (a) GGA spin-up, (b) GGA spin-down, (c) mBJ spin-up, (d) mBJ spin-down

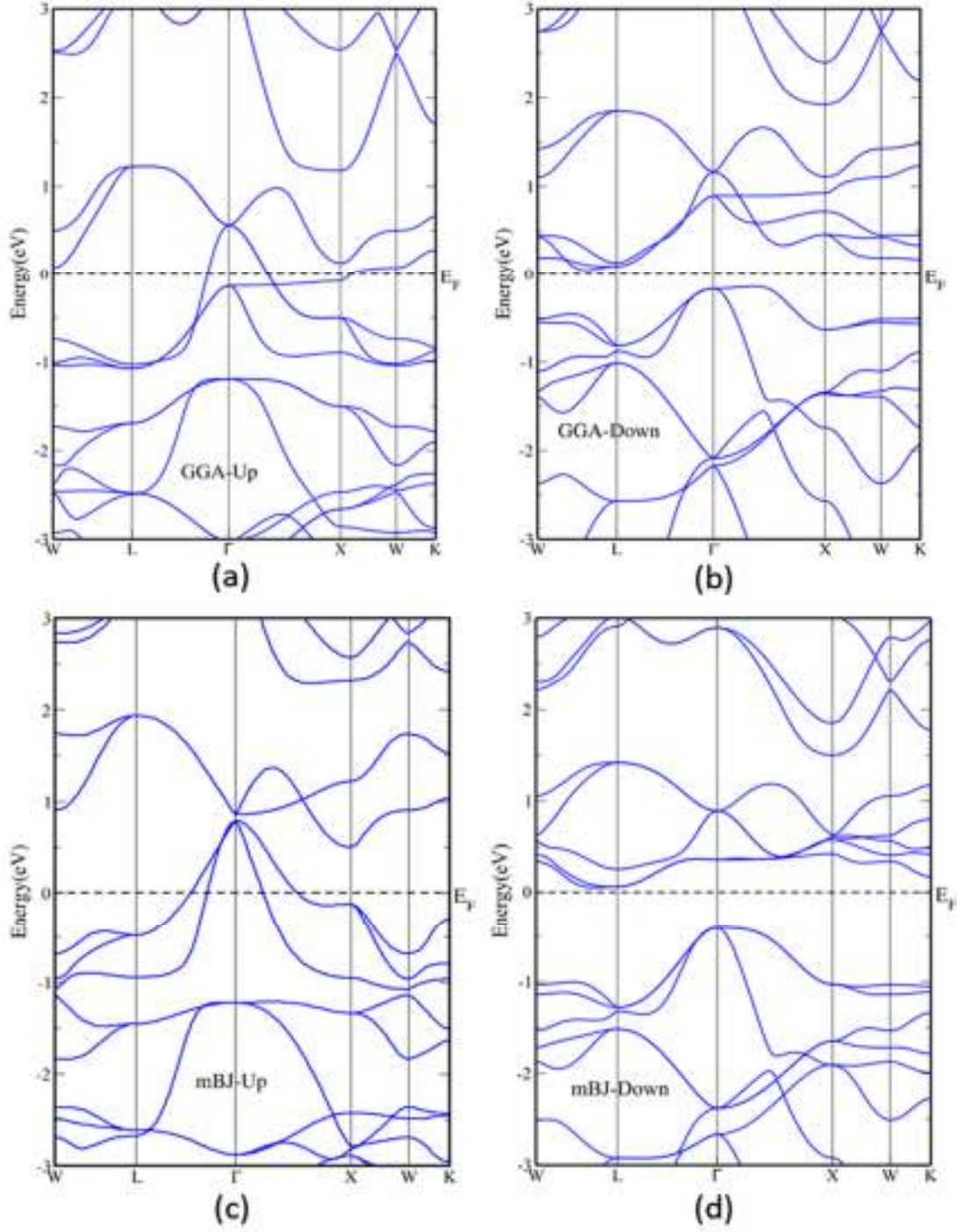


Figure A.37

2D charge density contours of FeMnScGa in the (110) plane for (a) majority spin, (b) minority spin

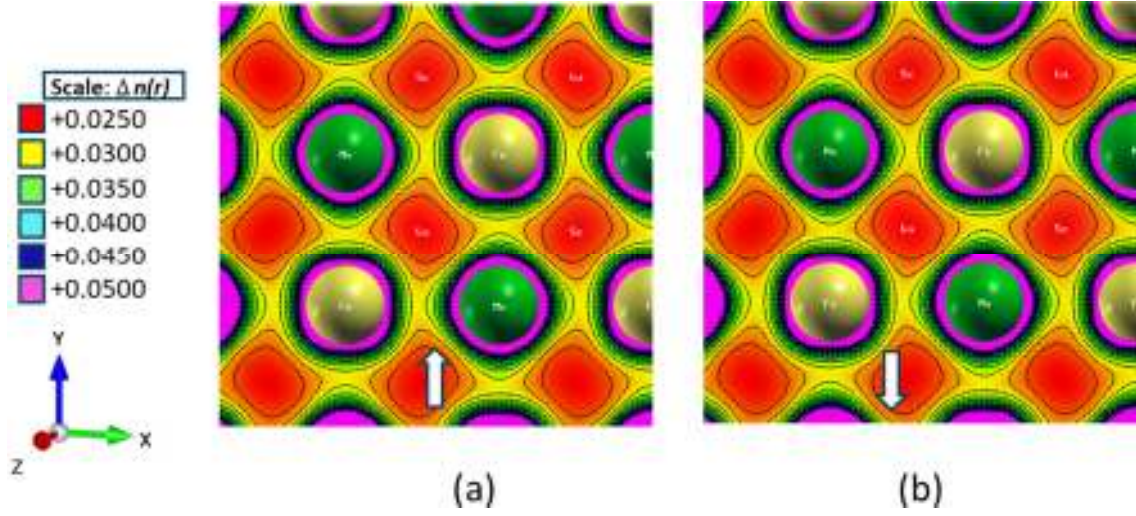


Figure A.38

Comparison of Energies and Stability for FM, PM, and AFM Configurations

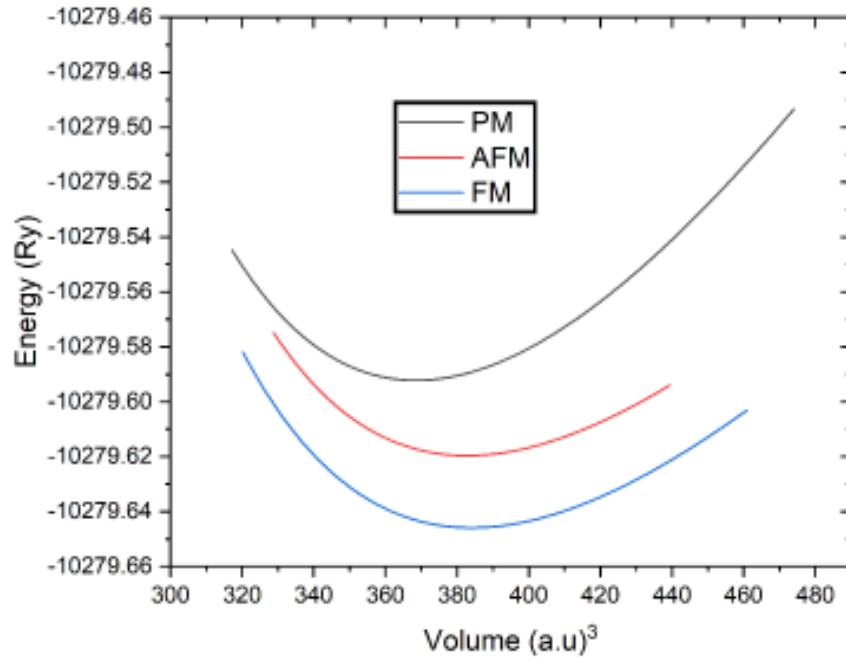


Figure A.39

(a-d) Spectroscopic analysis of the dielectric function of FeMnScGa (0-8 eV)

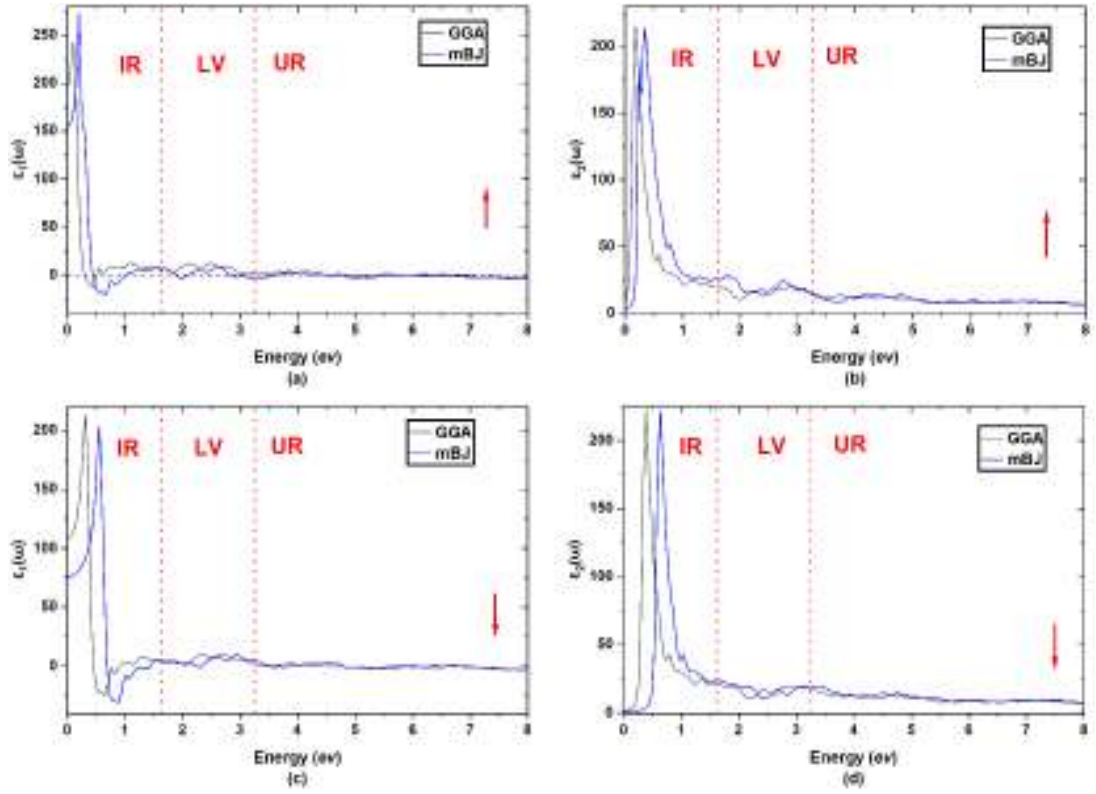


Figure A.40

(a-d) Refractive index and extinction coefficient of FeMnScGa for both spin states

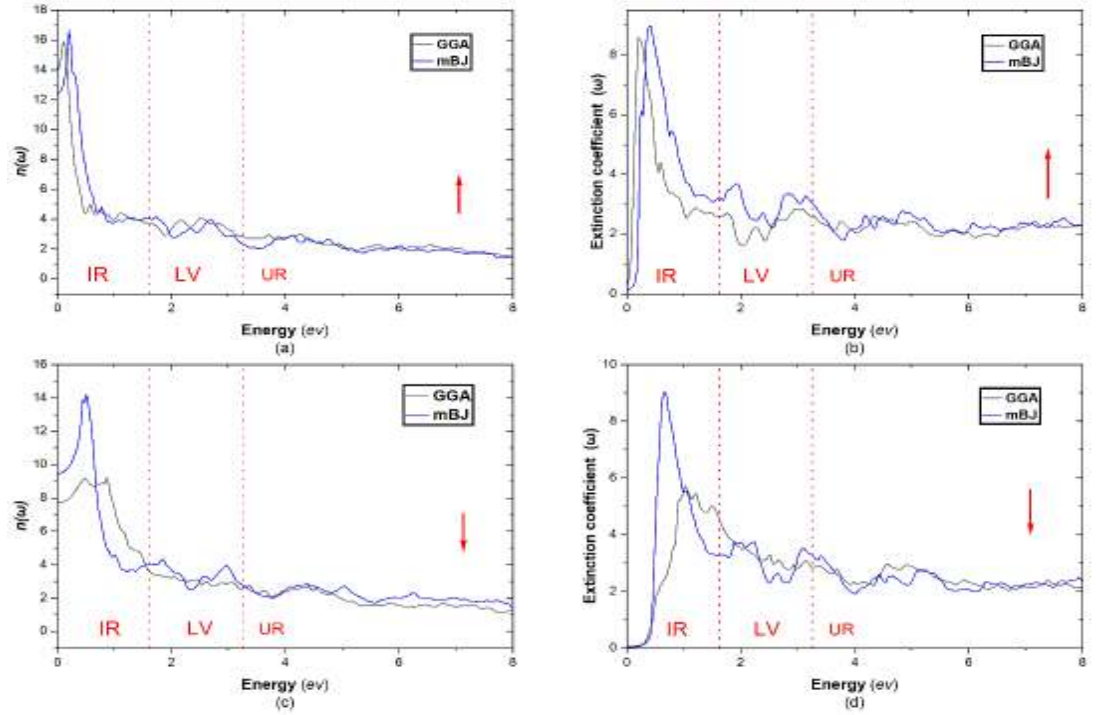


Figure A.41

Calculated Absorption Spectra of FeMnScGa (GGA and mBJ) for Both Spin States

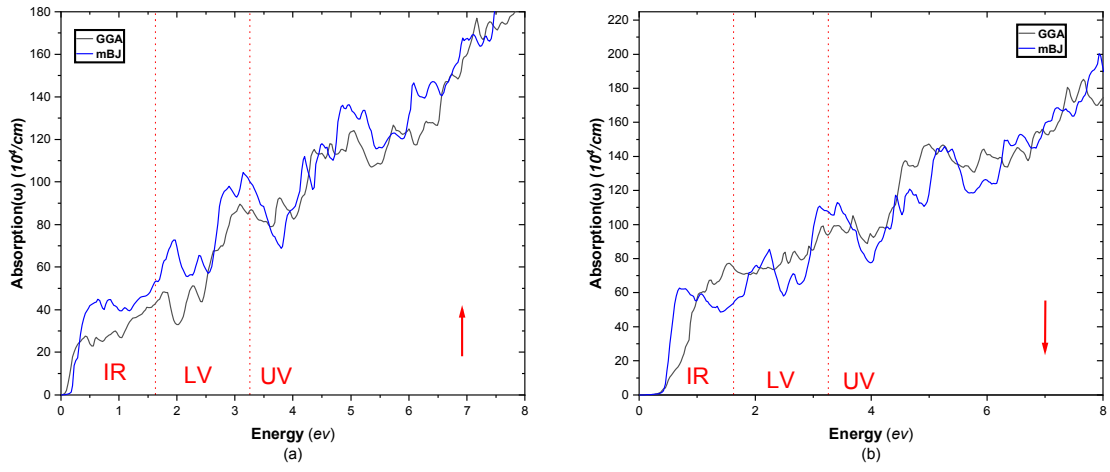


Figure A.42

Calculated optical conductivity of FeMnScGa (GGA and mBJ) for spin-up and spin-down states

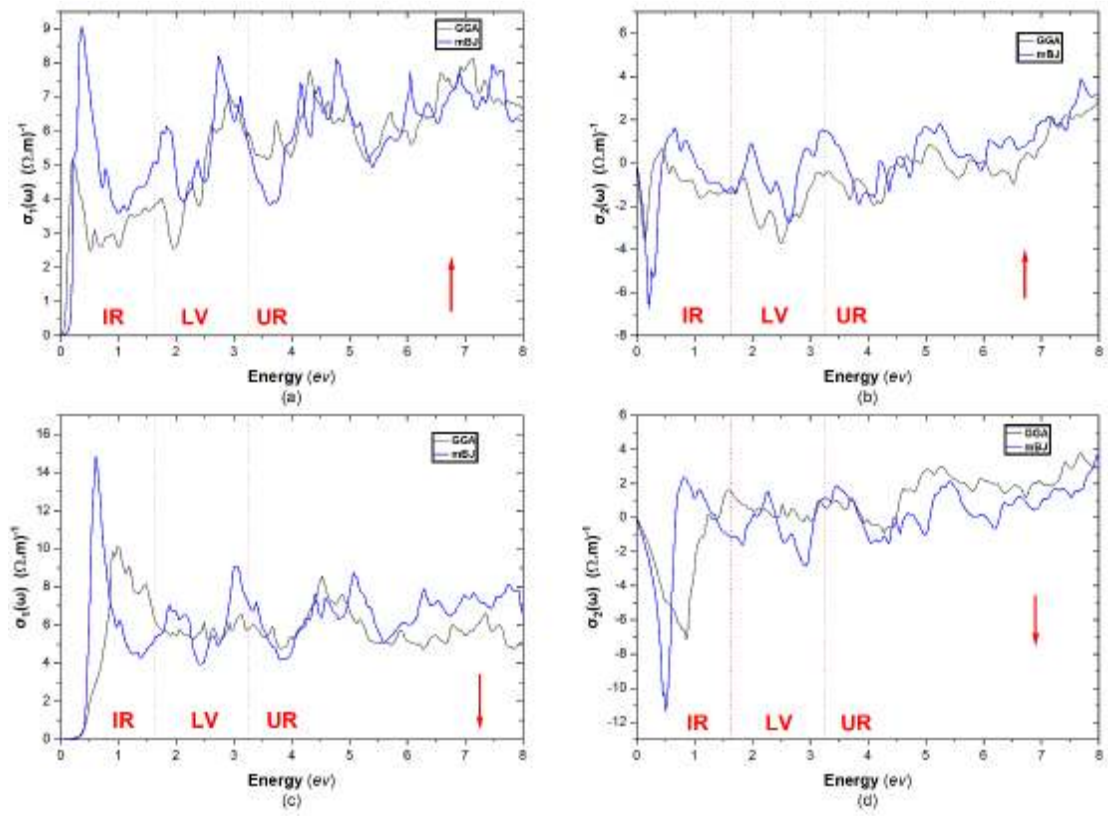


Figure A.43

(a) & (b) Reflectivity spectra of FeMnScGa were computed for spin-polarized states using both GGA and mBJ methods

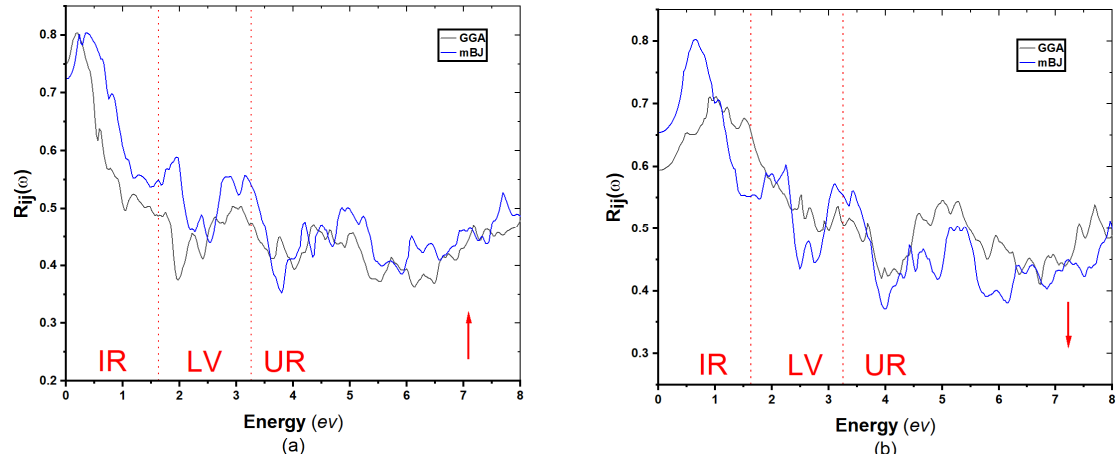


Figure A.44

Calculated energy loss function of FeMnScGa (GGA and mBJ) for spin-up and spin-down states

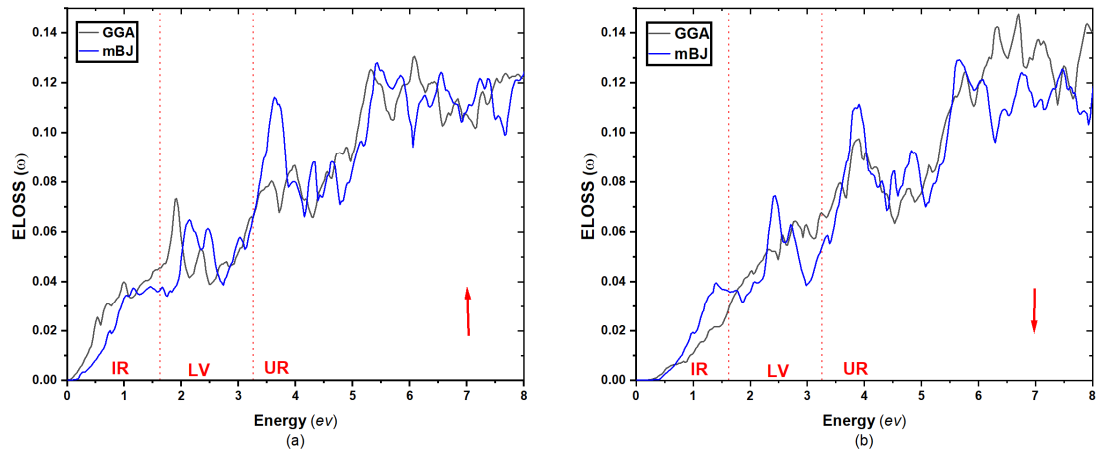


Figure A.45

Thermoelectric characteristics of FeMnScGa under varying temperature conditions

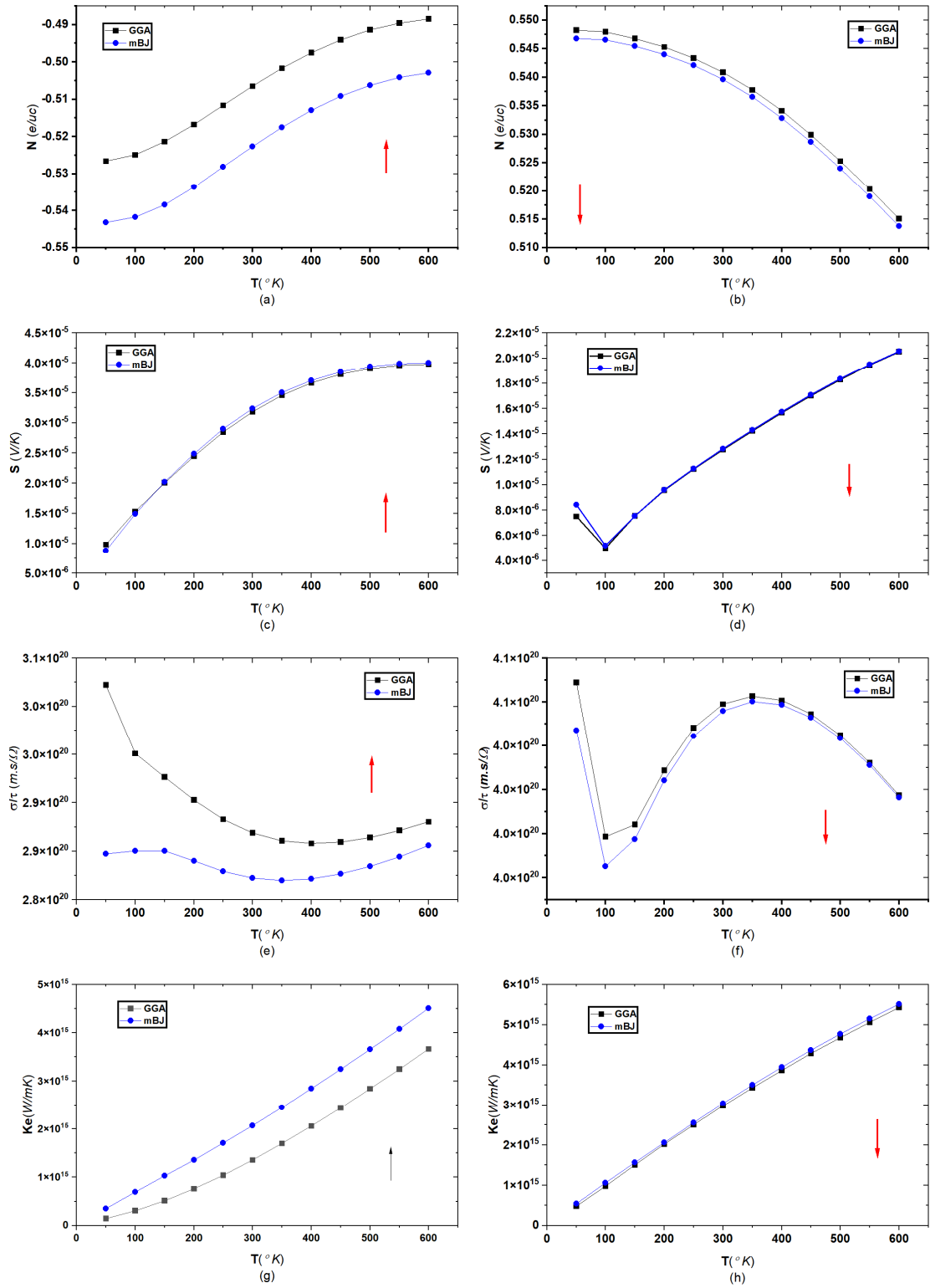


Figure A.46

Temperature dependence of spin-resolved ZT and PF in thermoelectric performance

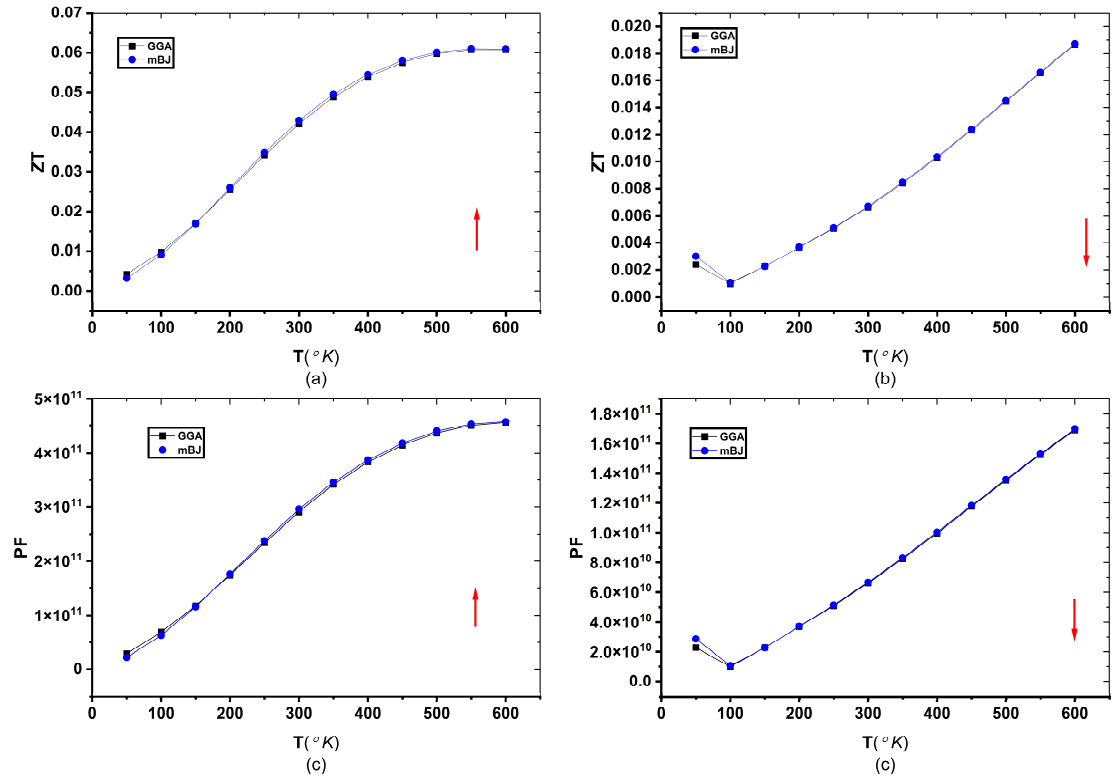


Figure A.47

(a) & (b) Hall Coefficient (R_H) for Spin-Up and Spin-Down Channels in FeMnScGa Using GGA and mBJ

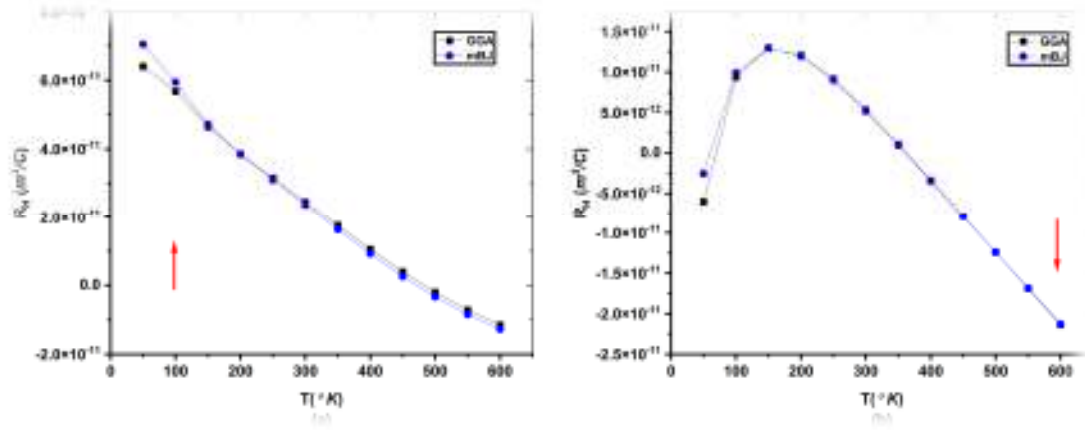


Figure A.48

Lattice Thermal Conductivity (κ_l) of FeMnScGa Alloy Across Temperatures Using GGA and mBJ

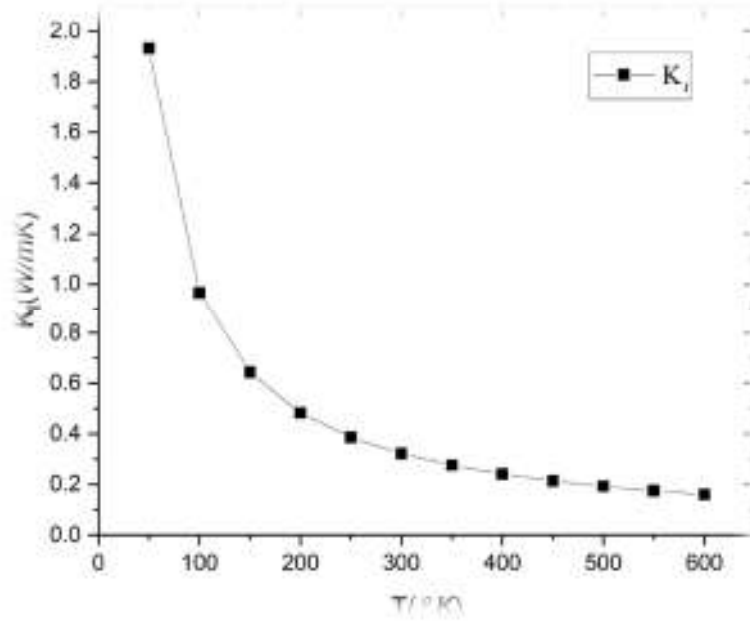
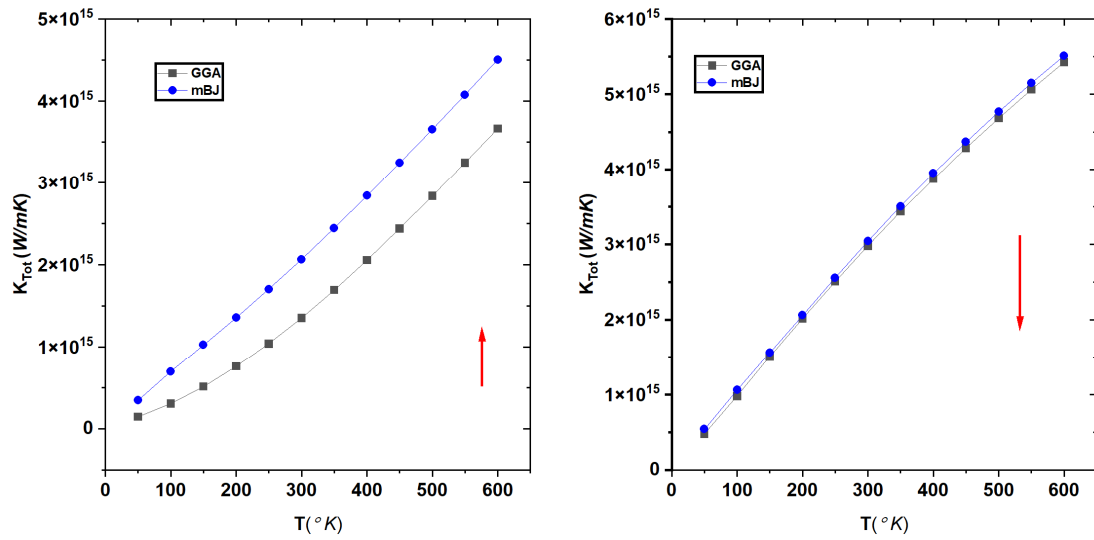


Figure A.49

Total thermal conductivity κ_l) of FeMnScGa alloy across temperatures using GGA and mBJ



Appendix B

Tables

Table B. 1

Temperature-dependent ZT and PF values for spin-up and spin-down configurations

Parameter	50K	50K	300K	300K	500K	500K
	(GGA)	(mBJ)	(GGA)	(mBJ)	(GGA)	(mBJ)
$ZT\uparrow$	0.02897	0.03057	0.01372	0.02024	0.04623	0.0559
$ZT\downarrow$	0.34034	0.54548	0.59639	0.62841	0.39418	0.40836
$PF\uparrow (\Omega msK)^{-1}$	5.19×10^{10}	5.09×10^{10}	3.07×10^{10}	4.68×10^{10}	1.40×10^{11}	1.75×10^{11}
$PF\downarrow (\Omega msK)^{-1}$	1.11×10^{11}	1.14×10^{11}	8.71×10^{11}	8.09×10^{11}	9.88×10^{11}	9.43×10^{11}

Table B.2

Hall Coefficient (R_H) values for FeMnScAl alloy for specific temperature values

	GGA	mBJ	GGA	mBJ
T(°K)	$R_H\uparrow$	$R_H\uparrow$	$R_H\downarrow$	$R_H\downarrow$
50	-1.95×10^{-9}	-2.24×10^{-9}	-7.58×10^{-9}	-1.77×10^{-8}
300	-2.01×10^{-9}	-1.83×10^{-9}	-1.72×10^{-9}	-2.34×10^{-9}
500	-1.38×10^{-9}	-1.20×10^{-9}	-7.29×10^{-9}	-9.20×10^{-10}

Table B.3

Structural and stability parameters of FeMnScGa

FeMnScGa	E_{GROUND} (Ry)	a (Å)	B (GPa)	B'	$E_{Form.}$ (Ry)	$E_{cohesive}$ (Ry)
Type (1)	-10279.621028	6.1555	104.3154	5.1878	-7.509	1.211102
Type (2)	-10279.645909	6.1061	118.2966	5.3287	-7.4895	1.235983
Type (3)	-10279.643407	6.2187	96.8569	4.0375	-7.5074	1.233481

Table B.4*Elastic Constants of FeMnScGa at Zero Strain*

Property	Value	Unit
C_{11}	171.4087	GPa
C_{12}	105.3026	GPa
C_{44}	87.9522	GPa
Bulk Modulus (B)	127.337	GPa
Shear Modulus (S_R)	52.843	GPa
Young's Modulus (Y)	139.264	GPa
Poisson's Ratio (ν)	0.31700	-
B/ S_R	2.40972	-
A	2.661	-
Average wave velocity ($v_{\square}$)	3384.18	m/s
Debye Temperature	413.495	K
Melting Temperature	1566.0254 $\pm$ 300	K

Table B.5*Partial and Total Magnetic Moments in FeMnScGa Alloy from GGA and mBJ Calculations*

Component	GGA (μ_B)	mBJ (μ_B)
Interstitial	-0.02987	-0.05478
Sphere (Fe)	0.31998	0.30333
Sphere (Mn)	2.91822	2.95105
Sphere (Sc)	-0.13953	-0.15085
Sphere (Ga)	-0.04926	-0.05687
Total Spin Magnetic	3.01954	2.99188

Table B.6*Total Energy and Structural Parameters for FM, PM, and AFM States for FeMnScGa alloy*

Magnetic State	(B) (GPa)	(B')	E_0 (Ry)	a ($\text{\AA}$)
AFM	120.8885	5.8671	-10279.61962	6.0986
PM	147.8004	4.3458	-10279.59223	6.0229
FM	5.3287	118.2966	-10279.645909	6.1061

Table B.7*Key Optical Property Values for FeMnScGa Alloy (GGA and mBJ)*

Property	Spin State	Method	Value	Units
Static Dielectric Constant ($\epsilon_1(0)$)	Down	GGA	108.42	-
Static Dielectric Constant ($\epsilon_1(0)$)	Down	mBJ	75.79	-
$\epsilon_1(\omega)$ Peak Position	Down	GGA	0.32	eV
$\epsilon_1(\omega)$ Peak Position	Down	mBJ	0.55	eV
$\epsilon_2(\omega)$ Peak Position	Down	GGA	0.39	eV
$\epsilon_2(\omega)$ Peak Position	Down	mBJ	0.64	eV
Static Refractive Index ($n(0)$)	Up	GGA	14.01	-
Static Refractive Index ($n(0)$)	Up	mBJ	12.42	-
$n(\omega)$ Peak Position	Up	GGA	0.15	eV
$n(\omega)$ Peak Position	Up	mBJ	0.204	eV
$n(\omega)$ Peak Value	Up	GGA	15.08	-
$n(\omega)$ Peak Value	Up	mBJ	16.644	-
Static Refractive Index ($n(0)$)	Down	GGA	7.72	-
Static Refractive Index ($n(0)$)	Down	mBJ	9.46	-
$n(\omega)$ Peak Position	Down	GGA	0.45	eV
$n(\omega)$ Peak Position	Down	mBJ	0.53	eV
$\alpha(\omega)$ Peak Value (UV region)	Up	GGA/mBJ	$\sim 180 \times 10^4$	cm^{-1}
$\alpha(\omega)$ Absorption Threshold	Down	GGA	0.189	eV
$\alpha(\omega)$ Absorption Threshold	Down	mBJ	0.442	eV
$\sigma_1(\omega)$ Peak Value	Up	GGA	5.21	$\Omega \cdot \text{m}^{-1}$
$\sigma_1(\omega)$ Peak Value	Up	mBJ	8.92	$\Omega \cdot \text{m}^{-1}$
$\sigma_1(\omega)$ Peak Position	Up	GGA	0.23	eV
$\sigma_1(\omega)$ Peak Position	Up	mBJ	0.37	eV
$\sigma_1(\omega)$ Threshold	Down	GGA	0.189	eV
$\sigma_1(\omega)$ Threshold	Down	mBJ	0.442	eV
$\sigma_1(\omega)$ Peak Value	Down	GGA	10.07	$\Omega \cdot \text{m}^{-1}$
$\sigma_1(\omega)$ Peak Value	Down	mBJ	14.86	$\Omega \cdot \text{m}^{-1}$
$\sigma_1(\omega)$ Peak Position	Down	GGA	0.612	eV
$\sigma_1(\omega)$ Peak Position	Down	mBJ	1.02	eV
Static Reflectivity ($R(0)$)	Up	GGA	75%	-
Static Reflectivity ($R(0)$)	Up	mBJ	72%	-
Reflectivity Peak ($R(\omega)$) (IR region)	Up	GGA/mBJ	80%	-
Static Reflectivity ($R(0)$)	Down	GGA	59%	-
Static Reflectivity ($R(0)$)	Down	mBJ	65%	-

Table B.8*Thermoelectric Properties of FeMnScGa Alloy*

Property	Spin State	50 K (GGA)	50 K (mBJ)	300 K (GGA)	300 K (mBJ)	600 K (GGA)	600 K (mBJ)
(N)	(↑)	-0.5266	-0.5432	-0.5065	-0.5227	-0.4885	-0.5029
	(↓)	0.54828	0.54677	0.54089	0.53962	0.5151	0.51382
(S) $\mu\text{V/K}$	(↑)	24.5	24.9	31.9	32.4	39.8	40.0
	(↓)	9.58	9.58	12.76	12.82	20.51	20.55
(σ/τ) $m\cdot s/\Omega\cdot eV$	(↑)	3.1×10^{20}	2.9×10^{20}	2.87×10^{20}	2.82×10^{20}	2.88×10^{20}	2.86×10^{20}
	(↓)	4.068×10^{20}	4.047×10^{20}	4.059×10^{20}	4.055×10^{20}	4.017×10^{20}	4.016×10^{20}
$(\kappa\Box)$	(↑)	1.48×10^{14}	3.47×10^{14}	3.67×10^{15}	4.51×10^{15}	3.67×10^{15}	4.51×10^{15}
	(↓)	5.42×10^{14}	1.06×10^{15}	3.04×10^{15}	3.50×10^{15}	5.51×10^{15}	4.51×10^{15}
(ZT)	(↑)	0.00417	0.00319	0.04213	0.04292	0.06076	0.06094
	(↓)	0.00239	0.00301	0.00666	0.00672	0.01868	0.01874
(PF)	(↑)	2.92×10^{10}	2.21×10^{10}	2.91×10^{11}	2.96×10^{11}	4.56×10^{11}	4.58×10^{11}
	(↓)	2.29×10^{10}	2.88×10^{10}	6.61×10^{10}	6.67×10^{10}	1.69×10^{11}	1.70×10^{11}
(R_H)	(↑)	6.40×10^{-11}	7.05×10^{-11}	2.46×10^{-11}	2.37×10^{-11}	-1.15×10^{-11}	-1.27×10^{-11}
	(↓)	-5.99×10^{-12}	-2.52×10^{-12}	-2.14×10^{-11}	-2.13×10^{-11}	-2.14×10^{-11}	-2.13×10^{-11}

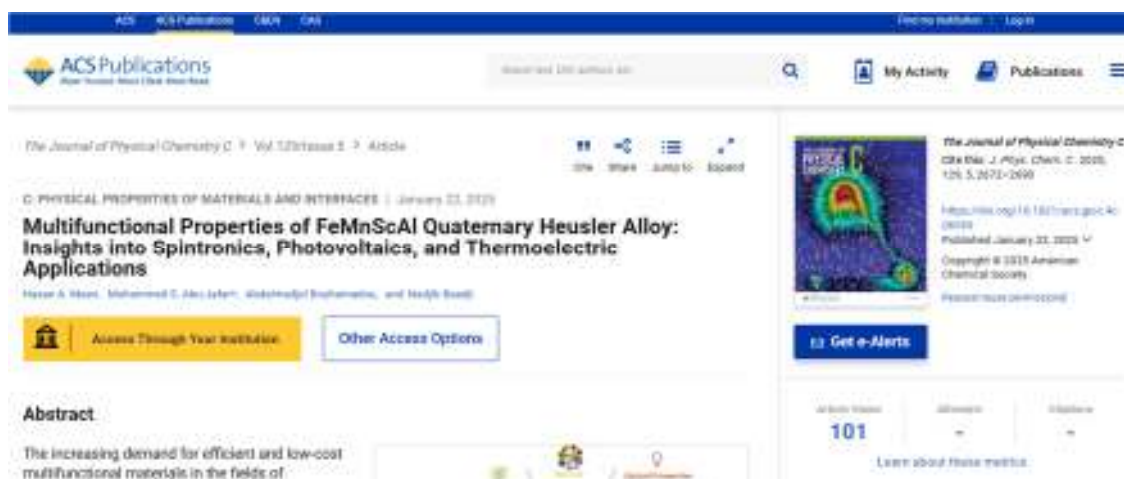
Table B.9*K-Point Convergence for FeMnCrGe, FeMnScAl, and FeMnScGa*

Compound	K-points	Total Energy (Ry)
FeMnCrGe	100	-11.36318153
	500	-11.49779171
	1000	-11.49601835
	2000	-11.49601349
	3000	-11.49600349
	4000	-11.49600349
FeMnScAl	100	-6877.044820
	500	-6877.045405
	1000	-6877.045866
	2000	-6877.045669
	3000	-6877.045673
	4000	-6877.045673
FeMnScGa	100	-10279.616838
	500	-10279.618146
	1000	-10279.619354
	3000	-10279.6218882
	4000	-10279.6218882

Appendix C

Certificates of acceptance of the research extracted from the dissertation

1. Multifunctional Properties of *FeMnScAl* Quaternary Heusler Alloy: Insights into Spintronics, Photovoltaics, and Thermoelectric Applications



The Journal of Physical Chemistry C

Impact Factor: 3.3

Ranked Q1

2. Extensive DFT study of *FeMnCrGe* quaternary Heusler alloy: structural, elastic, magnetic, optical and thermoelectric properties



Optical and Quantum Electronics

Impact Factor: 3.3

Ranked Q2



جامعة النجاح الوطنية
كلية الدراسات العليا

استكشاف الخصائص التركيبية والميكانيكية والإلكترونية
والمغناطيسية والبصرية والكهروحرارية لسبائك هزlr الرباعية
للمركبات FeMnScGa و FeMnScAl و FeCrMnGe

إعداد

حسن علي حسن مصري

إشراف

أ. د. محمد أبو جعفر

قدمت هذه الأطروحة استكمالاً لمتطلبات الحصول على درجة الدكتوراه في الفيزياء،
بكلية الدراسات العليا في جامعة النجاح الوطنية في نابلس - فلسطين.

استكشاف الخصائص التركيبية والميكانيكية والإلكترونية والمغناطيسية والبصرية والكهرباء الحرارية لسبائك هزلر الرباعية للمركبات FeMnScAl و FeMnScGa و FeMnCrGe

إعداد

حسن علي حسن مصري

إشراف

أ. د. محمد أبو جعفر

الملخص

تقدم هذه الأطروحة دراسة شاملة تعتمد على المبادئ الأولى لثلاث سبائك هزلر رباعية وهي FeMnScAl و FeMnScGa و FeMnCrGe، مع التركيز على خصائصها التركيبية، الإلكترونية، المغناطيسية، البصرية، والكهرباء الحرارية. تم استخدام حسابات نظرية الكثافة الوظيفية، مع تطبيق كل من تقريب GGA-PBE وإمكانات بيك-جونسون المعدلة (mBJ)، لتوفير رؤى مفصلة حول إمكانات هذه السبائك للتطبيقات في مجالات الإلكترونيات الدورانية وتحولات الطاقة.

تتبلور جميع المركبات الرباعية في تراكيب مكعبة متركزة الوجه مجموعة الفضاء (Fm-3m)، حيث أظهرت أكثر تكويناتها استقراراً طاقات تكوين سالبة، مستوفية معايير الاستقرار الميكانيكي. ومن الجدير بالذكر أن التكوين من النوع الأول لـ FeMnCrGe يظهر صلابة هيكلية فائقة، في حين أن التكوينات من النوع الثاني لـ FeMnScAl و FeMnScGa تظهر استقراراً مثالياً يتميز بطبيعة رابطة مختلطة أيونية-تساهمية. تؤكد حسابات الثوابت المرنة على الاستقرار الميكانيكي لجميع المركبات، مع ملاحظة سلوك ميكانيكي غير متجانس.

تكشف تحليلات التركيب الإلكتروني عن سلوك معدني نصف كامل ملحوظ في جميع المركبات، مع تحقيق استقطاب دوران بنسبة 100%. يظهر FeMnCrGe فجوة نطاق دوران لأسفل تبلغ 0.974

إلكترون فولت (mBJ)، بينما تعرض FeMnScAl و FeMnScGa فجوات تبلغ 0.1574 و 0.442 إلكترون فولت على التوالي. تتبع العزوم المغناطيسية قاعدة سليتر-بولينج، وتتراوح بين 1.00 بورمغنتون FeMnCrGe و 3.00 بورمغنتون FeMnScAl و FeMnScGa لكل وحدة صيغة. وتساهم التفاعلات المغناطيسية القوية بين ذرات المنغنيز والعناصر المجاورة في السلوك المغناطيسي الحديدي القوي.

يكشف تحليل الخصائص الحرارية الكهروحرارية عن إمكانات واعدة لتحويل الطاقة، حيث يحقق FeMnScAl قيمة ZT قصوى تبلغ 0.689 عند درجة حرارة 150 كلفن، بينما يصل FeMnCrGe إلى 0.42 عند 500 كلفن. تشير الخصائص البصرية إلى امتصاص قوي للأشعة فوق البنفسجية، مع إظهار FeMnScGa معاملات امتصاص تتجاوز 10×180 سم. كما تظهر المواد مؤشرات انكسار عالية وانتقالات بين النطاقات مهمة، مما يشير إلى إمكانات تطبيقها في الأجهزة البصرية الإلكترونية.

تعد هذه النتائج مؤشراً على أن هذه المواد تمتلك مقومات قوية تؤهلها للاستخدام في للأجهزة الإلكترونية الدورانية من الجيل القادم، وتطبيقات الطاقة الكهروحرارية، والأنظمة البصرية الإلكترونية. يجمعون بين المعدنية النصفية الفريدة، والخصائص المغناطيسية القوية، والأداء الحراري الكهربائي الفعال، مما يجعلها ذات صلة كبيرة لمعالجة التحديات التكنولوجية الحالية في مجالات الإلكترونيات وتحويل الطاقة. يوفر الإطار النظري الشامل الذي تم تطويره في هذه الدراسة أساساً قوياً للتحقيقات التجريبية المستقبلية والتطبيقات العملية لهذه المواد المتعددة الاستخدامات في التقنيات المتقدمة.

الكلمات المفتاحية: سبيكة هيوزلر رباعية؛ البنية الإلكترونية؛ الخصائص المغناطيسية؛ أنصاف الفلزات؛ الخصائص المرنة؛ الخصائص البصرية؛ الخصائص الحرارية الكهربائية؛ طريقة الموجة المستوية المعززة الخطية كاملة الجهد (FP-LAPW).