Contents

		page
Abstract		I
Acknowledgements		iii
Contents		v
List of Figures		vii
List of Tables		xi
List of Symbols		xiii
List of Abbreviations		xv
Chapter 1: INTRODUCTION		
1.1	Chalcogenides and their evolution	1
1.2	Cu-based Chalcogenides for Photovoltaic(s) and Thermoelectric(s)	2
1.3	Motivation	3
1.4	Objectives	3
1.5	Outline of the Thesis	3
Chapter 2: Theory and Literature Review		
2.1	Photovoltaic Effect	5
2.2	Current transport in p-n junction diode	5
	2.2.1 Under dark	5
	2.2.2 Under illumination	6
	2.2.3 Conversion efficiency	7
2.3	Schockley Queisser Limit	7
2.4	Solar Spectrum	7
2.5	*Charges in equilibrium and under illumination*	8
2.6	*Recombination Mechanism*	9
	2.6.1 Radiative Recombination	10
	2.6.2 Auger Recombination	10
	2.6.3 Shockley-Read-Hall (SRH) Recombination	10
2.7	*Charge Carrier Transport Equations*	11
2.8	*Absorption of Light and Transmission loss-mitigation*	12
2.9	*Thin-film Solar Cell*	13
2.10	*Quaternary Chalcogenide Solar Cell*	13
2.11	*Thermoelectric Motivation and Applications*	15
2.12	*Critical Features Required for Effective Figure-of-Merit*	16
	2.12.1 Optimizing carrier concentration	16
	2.12.2 Band structure dependence	17
	2.12.3 Low lattice thermal conductivity	18
2.13	*Quaternary Chalcogenides as Thermoelectric Devices*	19
Chapter 3: Theory and Computational Methods		23
3.1	Many-Body Problem	23
3.2	Density Functional Theory (DFT)	25
	3.2.1 The Hohenberg-Kohn Theorems and Kohn-Sham approach	26
3.3	Self-Consistent Field calculation	27
3.4	Exchange and correlation functionals	27
	3.4.1 Local Density Approximation	27
	3.4.2 Generalised gradient approximation (GGA)	28
	3.4.3 Hybrid Functional	28
3.5	Limitations of DFT	28
3.6	Phonon Dynamics	29
	3.6.1 Density Functional Perturbation Theory (DFPT)	29
3.7	Transport Phenomena in solids	29
	3.7.1 Boltzmann Transport Equation (BTE)	30
	3.7.1.1 Relaxation Time Approximation (RTA)	31
	3.7.1.2 The electronic parameters derived from the BTE	31

3.7.2 Scattering mechanism for electrons 32
3.7.2.1 Polar-optical Scattering 33
3.7.2.2 Acoustic phonon scattering 33
3.7.2.3 Impurities scattering 33
3.7.3. Lattice thermal conductivity 34
3.7.4. Lattice thermal conductivity (particle and Wave-like) 34
3.7.5. Scattering process in phonons 35
3.7.5.1 Phonon-Phonon scattering 35
3.7.5.2 Phonon Boundary scattering 36
3.7.5.3 Isotope Scattering 36
3.8 SCAPS Simulator 36

Chapter 4: Materials Prospects in $Cu_2HgSn(Se_xS_{1-x})_4$ 39
4.1 Introduction 39
4.2 Computational Details 40
4.3 Results and Discussion 40
4.3.1 Structural properties 40
4.3.2 Elastic Properties 42
4.3.3 Electronic properties 43
4.3.4 Lattice Parameter and Band gap bowing 44
4.3.5 Optical properties 45
4.3.6 Impact of bowing on effective mass and dielectric constant 46
4.4 Conclusion 47

Chapter 5: Single- / multiple- junction(s) and graded bandgap solar cell 49
5.1 Introduction 50
5.2 Device Modelling and Simulation 52
5.3 Single junction Device Analysis 52
5.3.1 Role of absorber CHTS layer thickness on photovoltaic response 52
5.3.2 Role of absorber CHTS layer doping concentration on solar cell 53
5.3.3 Role of defect concentration at absorber CHTS and CHTS/CdS interface 54
5.3.4 Role of Buffer layer 55
5.3.5 Role of back contact metal 57
5.4 Modelling Approach for Tandem Junction and Graded Band Gap Device 58
5.4.1 Tandem Structure 58
5.4.2 Graded Structure 59
5.5 Results and Discussions 60
5.5.1 Tandem Configuration 60
5.5.2 Graded Configuration 63
5.5.2.1 Significance of grading pattern 64
5.5.2.2 Significance of Positive Grading 65
5.5.2.3 Doping and Defect Density Effect 66
5.5.2.4 Hetero-interface band geometry effect 67
5.6 Conclusion 68

Chapter 6: Cu_2HgSnS_4: Phonon Dynamics & Thermoelectrics 71
6.1 Introduction 71
6.2 Computational Details 72
6.3 Results and Discussion 72
6.3.1 Structural and Electronic properties 72
6.3.2 Phonon properties and Lattice Thermal Conductivity 74
6.3.3 Transport properties 78
6.4 6.3.4 Figure of merit 81
Conclusion 82
Chapter 7: Conclusion and Future scope of work 85

Publications 87
References 89