

Contents

List of Figures	XI
List of Tables	XVII
List of Abbreviations	XIX
1 Introduction	1
1.1 Motivation	1
1.1.1 Outline of the Dissertation	3
2 Fundamentals	5
2.1 Solar Cells	5
2.1.1 Absorption and Recombination	6
2.1.2 Charge Carrier Separation and Collection	9
2.1.3 Basic Solar Cell Equations	11
2.1.4 Solar Radiation and Efficiency Limits of Solar Cells	12
2.2 Metal Halide Perovskites	14
2.2.1 Optoelectronic Properties of Metal Halide Perovskites	16
2.2.2 Solar Cell Architectures	20
2.2.3 Perovskite Deposition Techniques	22
3 Co-Evaporation of Metal Halide Perovskites	25
3.1 Historical Development of Co-Evaporated Perovskite Solar Cells	28
3.2 Processing Advantages of the Co-Evaporation Technique	31
3.3 Challenges in Co-Evaporated Perovskite Thin Film Growth	32
4 Experimental and Analytical Methodology	35
4.1 Materials	35
4.1.1 Materials used for Perovskite Depositions and Solar Cell Preparations . .	35
4.1.2 Methylammonium Iodide used in Chapter 5	36

4.2	Deposition Techniques and Solar Cell Preparation	36
4.2.1	Co-Evaporation Process	36
4.2.2	Solar Cell Architecture and Contact Layer Depositions	38
4.2.3	Monolithic Perovskite/Silicon Tandem Solar Cells	39
4.3	Characterization Methods	41
4.3.1	Solar Cell Characterization	41
4.3.2	Thin Film Characterization	44
4.3.3	Nuclear Magnetic Resonance Spectroscopy	45
4.3.4	Mass Spectrometry	45
5	Methylammonium Iodide: Influence of Impurities on Sublimation, Decomposition, and Condensation Dynamics	47
5.1	Introduction	48
5.2	Results and Discussion	51
5.2.1	Purity and Evaporation Behavior of Methylammonium Iodide	51
5.2.2	Mass Spectrometry of Decomposition Products	56
5.2.3	Reaction of Decomposition Products and Lead Iodide	60
5.3	Summary	66
6	Impact of Substrate Temperature and Hole-Transport Layer on Co-Evaporated Methylammonium Lead Iodide Solar Cells	69
6.1	Introduction	70
6.2	Results and Discussion	72
6.2.1	Pure Precursor Materials: Methylammonium Iodide and Lead Iodide	72
6.2.2	Co-Evaporation of Methylammonium Iodide and Lead Iodide	74
6.2.3	Substrate Temperature Limits for Co-Evaporation	77
6.2.4	Co-Evaporated Methylammonium Lead Iodide Solar Cells	80
6.2.5	Methylammonium Lead Iodide based Monolithic Perovskite-Silicon Tandem Solar Cells	84
6.3	Summary	88
7	Formamidinium Lead Iodide based Perovskites with High Stability	91
7.1	Introduction	91

7.2 Results and Discussion	94
7.2.1 Phase-stability of pure Formamidinium Lead Iodide Perovskites	94
7.2.2 Methylammonium Formamidinium Lead Iodide - Mixed Organic Cations for Stable Perovskite Solar Cells	101
7.2.3 Fully Textured Monolithic Perovskite/Silicon Tandem Solar Cells	111
7.3 Summary	118
8 Conclusions and Outlook	121
8.1 Methylammonium Iodide: Influence of Impurities on Sublimation, Decomposition, and Condensation Dynamics	122
8.2 Impact of Substrate Temperature and Hole-Transport Layer on Co-Evaporated Methylammonium Lead Iodide Solar Cells	123
8.3 Formamidinium Lead Iodide based Perovskites with High Stability	124
8.4 Outlook and Perspective	125
9 Bibliography	129
10 Appendix	159
10.1 Scientific Appendix to Chapter 5	159
10.2 Scientific Appendix to Chapter 6	162
10.3 Scientific Appendix to Chapter 7	164
List of Publications, Conference Contributions and Patents	173
10.1 Publications in Peer-Reviewed Journals	173
10.2 Conference Posters	175
10.3 Oral Conference Contributions	176
10.4 Patents	177
Supervised Students	178
Acknowledgments	179