In this thesis I shall present the most scientifically interesting and/or practically useful results achieved in my PhD project. Such results are related to fundamental properties and technological aspects of Cu2ZnSnS4 (CZTS) and related materials for solar cells. By "related materials" I mean two things: i) alternative solar absorbers (notably, Cu2SnS3) that are chemically related to CZTS and that have similar selling points; ii) other materials included in the device stack of CZTS solar cells. Here I list what I believe the main highlights of my work are.

First, we achieve the highest reported power conversion efficiency (5.2%) for a CZTS solar cell using pulsed laser deposition as a fabrication method for CZTS precursors. This is thanks to joint work of PhD student Andrea Cazzaniga, PhD student Chang Yan (University of New South Wales, Australia) and myself. Perhaps more importantly, we finally understand, albeit very roughly, the "rules of the game" for successful pulsed laser deposition of high-quality chalcogenide precursors for solar cells. This kind of understanding is not evident in the existing literature and is mostly the result of the work of PhD student Andrea Cazzaniga.

Second, I propose and test experimentally a modification of the standard CZTS solar cell architecture by inserting a very thin (few nm) CeO2 layer between the CZTS absorber and the CdS buffer. Despite being already known in the fields of catalysis and fuel cells, application of CeO2 in CZTS solar cells is completely new, even though the two materials have a nearly perfect lattice match. In a first investigation over a two-month external research stay at the University of New South Wales, I demonstrate that the open circuit voltage of standard CZTS solar cells fabricated by PhD student Chang Yan is boosted when I include a CeO2 interface passivation layer.

Third, I critically examine one of the mechanisms that are believed to be the major current issues of CTZS solar cells, namely recombination at the CZTS/CdS heterointerface. An initial outcome is a comprehensive review of the existing studies on the band alignment between the two materials, to which I add my own analysis and interpretation. I argue that, unlike what is often stated in the CZTS community, CdS does not necessarily have an unfavorable conduction band alignment with CZTS. Actually, the band alignment may to some extent be engineered by formation of secondary phases at the interface through controlled interdiffusion and due to orientation-dependent band alignment eects that are absent in the (otherwise very similar) Cu(In,Ga)Se2 solar cells. Another outcome of this sub-project is a collaboration with computational material scientists, mostly PhD student Mattias Palsgaard, to improve our theoretical understanding of the CZTS/CdS interface. A new computational method is applied to calculate some interface properties that are of interest but cannot be readily extracted by established methods. From a combination of atomistic- and device simulation it appears as if surface-state-induced band gap narrowing at the CZTS/CdS interface may be the main reason behind the poorer interface properties of CZTS/CdS solar cells compared to CZTSe/CdS solar cells. Interestingly, this problem may be solved by passivating those states with a Zn-based chalcogenide.

Fourth, I measure for the rst time the dielectric function of a monoclinic Cu2SnS3 thin film by spectroscopic ellipsometry. Cu2SnS3 is gaining some interest as a solar absorber and is produced by pulsed laser deposition by PhD student Rebecca Ettlinger. What is special about this study is the comparison with the dielectric function of Cu2SnS3 calculated from rst principles by external collaborators Rongzhen Chen and Clas Persson. We nd that the characteristic double absorption onset of monoclinic Cu2SnS3 is due to optical transitions from three closely spaced valence bands to a single conduction band. The different transitions are excited by diereent light polarization directions with respect to the crystal lattice, and this subtle distinction can only be resolved in the calculation when dense sampling in reciprocal space is employed.

Fifth, I undertake a comprehensive investigation of the properties of radio-frequency sputtered ZnO:Al thin lms used as a lateral electron transport layer on top of CZTS solar cells. With considerable fabrication help from M.Sc. student Tobias Ottsen, I demonstrate that compressive stress in the films is clearly correlated to several other properties (carrier concentration and mobility, grain size, orientation, and Al content) regardless of deposition pressure and position in the sputtering setup. Also, I show that spatial inhomogeneity in the electrical properties is mostly due to particle bombardment eects and only weakly to inhomogeneous oxygen distribution.

All the aforementioned results were achieved in the third and last year of my PhD project. In the rst two years of my project I established a CZTS solar cell device fabrication process ow at a university, and in a country, with zero experience with chalcogenide solar cells. The process includes deposition of all necessary thin lm materials in a CZTS device stack, CZTS excluded, and has so far resulted in 2.6% ecient in-house CZTS solar cells by pulsed laser deposition.

Throughout this thesis I try, when possible, to connect the physics and chemistry of the individual component materials to the resulting device physics, since the existing literature of CZTS solar cells is heavily biased toward the former aspect. The relationship between the two aspects is one of my main research interests.