The photophysics of solution processable semiconductors for applications in optoelectronic devices

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Summary

Solar energy has the potential to make a great contribution towards solving the growing demand for energy worldwide – due to its' being both renewable and relatively well distributed over the globe. It has gained much traction over the past few decades via increased research and development efforts, as well as from increased international attention to climate change, global warming and the need to limit pollution and engender a sustainable future. Traditionally, although silicon solar cells have been (and remain) the most purchased solar cells over the past few decades and currently have module-scale efficiencies exceeding 20%; emerging solar cell technologies (based on materials such as organics, colloidal quantum dots and perovskites) have become increasing popular and continuously challenge the dominance of silicon. This is especially true for niche applications where their flexibility, lightweight and wide range of colors give them advantages over silicon. In particular, this enables coating ultrathin layers that conform to the underlying substrate and makes them ideal for technologies such as disposable electronics, sensors, semi-transparent barriers, wearables and even in (external) facades of buildings where aesthetics are important. Although the economic barrier to widespread commercialization (in terms of the USD per Watt levelised cost criterion) has still not been met by most of these emerging technologies, positioning them as candidates for either niche applications (as above) or as efficient sub-cells for tandem architectures opens a much wider market for them than the already silicon-dominated solar cell market.

This thesis begins with an introduction to the different types of solution processable semiconductors studied and addresses their most relevant properties for the topics covered followed by a short discussion of important concepts and methodologies for the characterizations carried out in later the thesis.

Chapter 2 deals with an approach of making a 3-component (ternary) blend of a polymer, fullerene derivative and lead sulphide quantum dots (PbS QDs) in order to increase the dielectric constant of the blend. The dielectric constant of blends for organic solar cells is believed to be one of several limiting factors to achieving proper exciton dissociation and thus, improved efficiency. Although the idea of ternary blends for organic photovoltaics (OPV) is not new, the main idea has always been to improve coverage of the solar irradiance spectrum by incorporating materials with complementary absorption rather than aiming to use the third component as a means to increase the dielectric constant of the blends. In the chapter, we add small amounts of PbS QDs to a blend of a narrow bandgap copolymer and a fullerene derivative and use the photoluminescence of the

interfacial charge transfer state as a measure of the local dielectric constant of the blend. The results of this chapter strongly point towards a reduction of the initial charge transfer state population upon addition of PbS QDs, which we interpret as evidence of a locally increased dielectric constant of the ternary blend.

In chapter 3, an alternative approach to OPV: the concept of ferroelectric OPV (FE-OPV), is revisited with the aim of reconciling disparate views in the literature. Therein, a newly developed (and previously unreported) semiconducting-ferroelectric block copolymer is used as a compatibilizer to overcome the severe phase segregation that often occurs when ferroelectric polymers are mixed with semiconducting ones in a ternary blend. The copolymer containing films obtained by our optimized recipe were smooth and pinhole free which enabled us to eliminate the effect of a suboptimal morphology; and to conclude that our observations underlie previous assertions that ferroelectric compensation in the blends effectively cancels out the dipolar alignment in the ferroelectric rendering the FE-semiconductor blend strategy for FE-OPV inoperative.

Yet another approach in the field of emerging PV materials over the past decade has been a move towards hybrid organic-inorganic material systems. These systems retain the ease of solution processability and are generally more robust in terms of operational stability and efficiency compared to organics. In Chapter 4, such a material system: hybrid perovskites, are introduced and the effect of both thin film microstructure and electron extraction layers on the performance of hybrid perovskite solar cells (HPSCs) are studied; with particular emphasis on the so-called light-soaking effect. The light soaking effect is a reversible increase in the performance of perovskite solar cells upon extended illumination, this temporal instability is a challenge that must be overcome if HPSC technology is to become commercially viable. The results of this chapter clarify the roles either a compact or coarse microstructure play in device performance and also the role of the electron extraction layer on the light soaking phenomenon; thus advancing our understanding of HPSCs and how to improve their performance.

Finally, in chapter 5, another hybrid organic-inorganic system consisting of perovskite shelled PbS QDs is studied. Therein a detailed photophysical characterization of bismuth-perovskite shelled PbS QDs is performed. Perovskite and perovskite-shelled QDs have recently become forerunners for several optoelectronic applications and are likely going to remain technologically relevant in the years to come. To this end, temperature- and power- dependent

photoluminescence (PL) measurements were used to characterize thin films made from an ink based on Bismuth-perovskite shelled PbS QDs. Our experiments revealed the existence of radiative below bandgap states at low temperatures, reminiscent of other previously studied ligands. In addition, we show that the PL efficiency of the oleic acid (OA) -capped sample is much higher than the Bismuth-shelled ones – which suffer from traps; most likely formed at their surfaces during the phase-transfer ligand exchange process. Nevertheless, measurements on FETs show the successful removal of the native OA ligands, displaying electron dominated transport with mobilities comparable to reported values for perovskite-shelled PbS-QDs; which have been extensively studied in literature.

In conclusion, this thesis presents the results of experiments on different classes of solution-processable optoelectronic materials (polymers, perovskites and QDs). It also addresses methodologies such as the use of QD additives to improve the dielectric constant of organic photovoltaic blends, the (related) *FE-OPV* strategy of using the intrinsic dipolar alignment of a ferroelectric polymer to enhance the PCE of OPVs, topics regarding hybrid organic-inorganic photovoltaic technologies such as the light-soaking effect in perovskite solar cells and the photophysical properties of perovskite shelled PbS QDs. The final results herein improve our understanding of these classes of solution-processable semiconductors and points towards avenues for further material (system) improvements.