PhD Thesis Defense DAVID SO 'Copper Indium Sulfide Colloidal Quantum Dot Solar Cells'

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Wednesday October 19, 11:00 h. ICFO Auditorium

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Functional Optoelectronics Nanomaterials

ICFO-The Institute of Photonic Sciences

The ubiquity of solar energy conversion technology is an exciting target which we aspire for in this century. Colloidal quantum dot (CQD) solar cells are an attractive platform, being low-cost and allowing facile control on film properties and device fabrication, but to date are dominated by PbS. $CuInS_2$ (CIS) is a non-toxic alternative, showing promise in the bulk and as sensitizer but has not been thoroughly investigated for solid-state CQD solar cells. In this work we aim to incorporate CIS nanocrystals (NCs) into the gamut of CQD solar cell materials by making optoelectronic CQD solids, by fabricating functional devices in various architectures and by identifying and overcoming the limiting material properties and device mechanisms.

We described a synthethic scheme that resulted in CIS NCs that canbe deposited as CQD solids with controllable thicknesses. CIS NCs are typically synthesized using long chain thiols which are difficult to remove from the NC surface. We introduced long chain amines and phosphines, decomposable chalcogenides and zinc oleate treatments to substitute the functions that the thiol performed. This led to cubic nanocrystals with controllable size, from 2 nm to 3.2 nm, and optical properties with bandgaps ranging from 2.5 eV to 1.5 eV. These particles are also copper poor, decorated with indium and zinc on the surface. With this, we were able to design a ligand exchange protocol using hard ligands for making solid-state CQD films which is a step forward to engineering optoelectronic devices.

We fabricated optoelectronic devices using these films and identified thin film properties that could limit device performance. These assemblies of CIS NCs couple to each other as seen in changes in photoluminescence lifetimes with distance. When the distance between nanocrystals is reduced, these p-type films conduct although having low mobilities (~10^{-5} $cm^2V^{-1}s^{-1}$), displaying features that signify high trap densities, such as high responsivity and long photoconductive decays. The ability to form solid-state films has allowed us to fabricate optoelectronic devices such as transistors, photodetectors and solar cells. In solar cells, CIS CQD solids have led to good open-circuit volages (VOCs), around 0.6 V, but low short-circuit currents (JSCs), ~1 mA/cm^2, fill factors (FFs), ~0.30. With the absorption coefficient of the CIS CQD solids, we found that the internal quantum efficiency (IQE) of these devices, is far below 10%. These highlight the importance of addressing the high trap density in the quantum dot solid.

We addressed the low JSCs and FFs in CIS CQD solar cells by utilizing a bulk heterojunction (BHJ) architecture. This non-annealed architecture was fabricated by increasing the pore size of the TiO_2 network which allowed for the uniform and deep infiltration of CIS NCs. By using a BHJ architecture, we improved the performance of CIS CQD solar cells: mainly from an increase in JSC and FF leading to a six-fold increase in efficiency from initially at 0.15% to 1.16%. From analysis of Suns-Voc, -Jsc and transient Voc and Jsc measurements, we have identified that BHJ devices have less trap-assisted recombination and a lower activation energy (EU) for hole extraction. This was confirmed by shifts in the full device photoluminescence. We suggested that the BHJ structure allows for quenching of deeper tail states near the valence band in CIS by the transfer of electrons from TiO_2.

In this work, we have started from synthetic molecular precursors, to engineering supramolecular structures in trying to introduce CIS NCs into CQD optoelectronics, showcasing various devices: field effect transistors, photodetectors, bilayer and BHJ solar cells. In each stage, we have highlighted material, film and device properties which will be necessary for good performance. Ultimately this work aims to stimulate new interest in further development of CIS CQD solar cells, opening the possibility for non-toxic CQD photovoltaics.

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