Colloidal Quantum Dot Solids for Photovoltaics: Doping Control and New Device Architectures

David Zhitomirsky[1], Huan Liu[2], Jiang Tang[3], Sjoerd Hoogland[1], Oleksandr Voznyy[1], Xihua Wang[4], Melissa Furukawa[1], Larissa Levina[1], Philipp Stadler[1], Zhijun Ning[1], Illan Kramer[1], Edward H. Sargent[1]

[1]Electrical and Computer Engineering University of Toronto, 27 King's College Circle, Toronto, Ontario, Canada M5S 1A1
[2]School of Optical and Electronic Information, Huazhong University of Science and Technology, 1037 Luoyu Rd., Wuhan, Hubei 430074, China
[3]Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, 1037 Luoyu Rd., Wuhan, Hubei 430074, China
[4]Electrical and Computer Engineering University of Alberta, 116 St. and 85 Ave., Edmonton, AB, Canada T6G 2R3.

Colloidal quantum dots (CQDs) combine facile solution processing with bandgap tuning quantum effects, and are promising materials for inexpensive and highly efficient solar cells and other optoelectronic devices. The current approach to fabricate CQD photovoltaic devices employing a p-n junction has relied on combining a conductive, bandgap tunable p-type PbS CQD solid with another fixed bandgap n-type material, e.g. titanium dioxide (TiO2). Though efficient for a very narrow range of CQD bandgaps centered about 1.3 eV, this scheme falls short when bandgaps optimal for multiple junction implementations are considered, due to lack of bandgap control in the bulk semiconductor.

We report a novel approach to creating n- and p-type materials within the same PbS CQD material system, which are stable, chemically compatible, high performance in a photovoltaic context, and quantum confined on each side of the junction. The n-type materials are engineered by employing a halogen passivation approach in an air free environement, as oxygen is a common p-type dopant in these materials. P-type materials are obtained by working in air ambient and using silver doped PbS CQDs. We verify the doping type, magnitude, and mobility of these materials by using field effect transistors. We find that doping in CQDs can be explained by the stoichiometry of the overall film obtained through X-Ray Photoelectron Spectroscopy or Rutherford Backscattering - the sum of atomic oxidation number contributions dictates whether a material will be n-type or p-type.

We achieve a broad doping spectrum (10^{16}-10^{18} cm^{-3}) for both types of quanutm solid, allowing us to combine these materials into a functional photovoltaic device. A heavily doped (>10^{18} cm^{-3}) p-type CQD layer is interfaced with a lighter doped (10^{16} cm^{-3}) n-type CQD layer, to yield a compatible and functional rectifying junction. Such quantum-to-quantum junctions can span a range of bandgaps from 0.6 eV to 1.6 eV, which are highly desirable for multiple junction solar implementations that would otherwise be unattainable with current nanocrystal-to-bulk p-n interfaces.

We then compared this new approach to the conventional CQD solar cell architectures by building devices with different bandgaps. In the case of the 0.6 eV bandgap device in particular, 80% of the theoretical open circuit voltage limit was achieved, whereas a functional device could not be constructed using the conventional CQD-to-Bulk scheme with this same choice of bandgap. Under simulated AM1.5G illumination at 25°C, an optimal single quantum junction device with a 1.3 eV bandgap, showed a short-circuit current density J_{sc} of 20.9 mA/cm^2, an open-circuit voltage V_{oc} of 0.54 V, and a fill factor of 49%, resulting in a solar power conversion efficiency of 5.5%, already comparable with present state-of-the-art devices. Furthermore, these devices exhibit excellent thermal stability at 90°C and excellent performance retention over a span of 60 hours.

Control over the majority carrier within the same CQD material system, and the successful combination of these materials into a single device, offers a new engineering avenue for quantum dot optoelectronic devices.