Biexciton Dissociation Efficiency at Quantum Dot-Oxide Interfaces

MISCHA BONN, HAI WANG, ENRIQUE CANOVAS, Max Planck for Polymer Research — Harvesting multiexcitons populating semiconductor quantum dots (generated by carrier multiplication, CM) has been proposed as a path towards higher efficiencies in photovoltaic devices. Although CM efficiency has been widely interrogated in colloidal QD solutions, less focus has been placed on the physics regarding biexciton collection at electrodes. We investigate interfacial biexciton transfer dynamics from PbS quantum dots directly nucleated onto mesoporous SnO$_2$ films as a function of impinging photon flux and photon energy. A priori, this system seems very well-suited for achieving efficient biexciton dissociation, as the ultrafast QD-to-oxide transfer rate for 800nm excitation is substantially faster than Auger relaxation. Remarkably, the biexciton dissociation efficiency is below the detection efficiency, i.e. essentially zero. This seemingly counterintuitive result can be understood by noting that efficient hot electron transfer at the QD-oxide interface can compete with CM within the QDs. Hot electron transfer is observed to occur on sub-100 fs timescales, nulling the CM efficiency. Implications of these results for solar energy conversion are discussed.

Mischa Bonn
Max Planck for Polymer Research

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