Relaxation dynamics of photogenerated electrons and holes in quantum dots and applications to photon conversion in quantum dot solar cells

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Femtosecond transient absorption (TA) spectroscopy has been used to characterize charge carrier relaxation from the second excited state (1P) to the first excited state (1S) in colloidal indium phosphide (InP) quantum dots (QDs). A three-pulse experiment consisting of a visible pump, infrared pump, and white light probe, was used to characterize the relaxation of photo-generated excitons, and the roles of surface chemistry and size were investigated. A two-pulse experiment consisting of only the infrared pump and white light probe was used to characterize the relaxation of chemically injected electrons in the absence of holes. In the case of photo-generated excitons, two subsets of QDs are probed in the experiment, corresponding to exciton-confined and charge-separated QDs. The relaxation rates for exciton-confined and charge-separated QDs increase with decreasing QD diameter. The relaxation rates obtained for both photo-generated excitons that become charge-separated, and chemically injected electrons are slowed by approximately one order of magnitude compared to excitons confined to the QD core. Theoretical calculations of the electronic structure and expected optical transitions in InP QDs based on an atomistic pseudopotential model are presented and compared to experimental results. The role of carrier dynamics in the performance of quantum dot solar cells for photon conversion are discussed.