Quantum Dot Sensitization of TiO_2 Crystals

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Multiple exciton generation, the creation of two electron hole pairs from one high-energy photon, is well established in bulk semiconductors, but assessments of the efficiency of this effect remain controversial in quantum confined systems like semiconductor nanocrystals or quantum dots (QDs). We initially used CdSe QDs and CdSe/ZnS core shell QDs to establish the methods to produce reproducible covalently attached monolayers of QDs on TiO_2 single crystals. We then moved on to PbS nanocrystals to demonstrate the collection of photocurrents with quantum yields greater than one electron per photon. The strong electronic coupling and favorable energy level alignment between PbS nanocrystals and bulk TiO_2 facilitate extraction of multiple excitons more quickly than they recombine as well as collection of hot electrons from higher QD states. Our results have implications for increasing the efficiency of photovoltaic devices by avoiding losses due to the thermalization of photogenerated carriers.