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Charge Transfer across Quantum Dot-Oxide Interfaces for High-Efficiency Photovoltaics

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Metal oxides constitute robust and relatively cheap semiconductor materials that are finding increasing applications in opto-electronics, but their band gaps are typically prohibitively wide for the generation of free charges through the absorption of visible light. Several approaches have been developed to circumvent this drawback. Specifically, the sensitization of mesoporous oxides by semiconductor quantum dot (QD) nanocrystals represents a promising route for the development of low-cost photovoltaics in QD sensitized solar cells. In addition to their tuneable band gap, QDs have the ability to generate multiple charge carriers from single photons by a process called carrier multiplication (CM), which potentially provides a means towards high-efficiency photovoltaics. Although CM has been widely interrogated in colloidal QDs in solution, the collection of those multiple charge carriers at oxide electrodes has not been clearly elucidated. The contribution of CM towards the overall device performance is ultimately determined by a competition between transfer to the electrode material and charge recombination within the QDs. We report interfacial electron transfer dynamics from quantum dots grown directly onto mesoporous oxide films. Such systems are well-suited for achieving efficient multiple charge transfer by CM, as electron transfer from QD-to-oxide is substantially faster than charge recombination. However, despite CM occurring in the QD, only one electron is transferred to the oxide. 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.