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Charge Transfer Dynamics in Semiconductor Quantum Dots Relevant to Solar Hydrogen Production.

TODD KRAUSS, University of Rochester

Artificial conversion of sunlight to chemical fuels has attracted attention for several decades as a potential source of clean, renewable energy. For example, in light-driven proton reduction to molecular hydrogen, a light-absorbing molecule (the photosensitizer) rapidly transfers a photoexcited electron to a catalyst for reducing protons. We recently found that CdSe quantum dots (QDs) and simple aqueous Ni²⁺ salts in the presence of a sacrificial electron donor form a highly efficient, active, and robust system for photochemical reduction of protons to molecular hydrogen. To understand why this system has such extraordinary catalytic behavior, ultrafast transient absorption (TA) spectroscopy studies of electron transfer (ET) processes from the QDs to the Ni catalysts were performed. CdSe QDs transfer photoexcited electrons to a Ni-dihydrolipoic acid (Ni-DHLA) catalyst complex extremely fast and with high efficiency. Even under high fluence, the relative fraction of TA signal due to ET remains well over 80%, and depopulation from exciton-exciton annihilation is minimal (6%). We also found that increasing QD size and/or shelling the core CdSe QDs with a shell of CdS slowed the ET rate, in agreement with the relative efficiency of photochemical H₂ generation. The extremely fast ET provides a fundamental explanation for the exceptional photocatalytic H₂ activity of the CdSe QD/Ni-DHLA system and guides new directions for further improvements.