Supramolecular Photochemistry 2.0: New Frontiers in Triplet Sensitization

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The study of sensitized triplet-triplet annihilation (TTA) processes that convert low energy photons to higher energy light or chemical products has reached maturity.^[1,2,3] In this process, selective excitation of long-wavelength absorbing triplet sensitizers in the presence of appropriate molecular acceptors enables TTA, resulting in either frequency upconverted light or the formation of desired chemical products. Various combinations of donor and acceptor have been explored and data will be presented on a number of these compositions spanning light conversions ranging from the near-UV to the near-IR. This presentation will also detail recent examples of photochemical transformations sensitized through the TTA process. Semiconductor (SC) nanocrystals are demonstrated to sensitize interfacial triplet-triplet energy transfer with molecular acceptors, providing a general paradigm for extracting triplet exciton energy from SC nanomaterials while extending their reactivity time by six orders-of-magnitude.^[4] The concept of SC nanocrystals serving as effective surrogates for molecular triplets suggests myriad of possible chemical and redox transformations relevant for fields as diverse as optoelectronics, solar energy conversion, and photobiology, as depicted in Fig. 1.

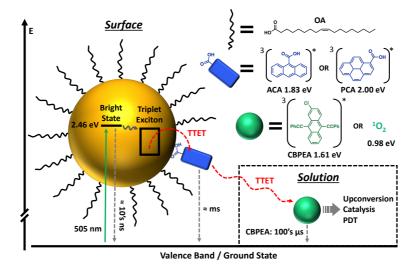


Figure 1. Supramolecular energy transduction at the semiconductor nanomaterial-molecular interface. Triplet excitons photogenerated in the nanomaterial are extracted as molecular triplet excited states bound to the surface that engage in subsequent excited state chemistry reactions with other partners freely diffusing in the solution.

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