Third Generation Solar Photon Conversion Based on Multiple Exciton Generation in Semiconductor Quantum Dots

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In order to utilize solar power for the production of electricity and fuel on a massive scale, it will be necessary to develop solar photon conversion systems that have an appropriate combination of high efficiency (delivered watts/m^2) and low capital cost ($\$/m^2$). One potential, long-term approach to high efficiency is to utilize the unique properties of quantum dot nanostructures to control the relaxation dynamics of photogenerated carriers to produce either enhanced photocurrent through efficient photogenerated electron-hole pair multiplication or enhanced photopotential through hot electron transport and transfer processes. To achieve these desirable effects it is necessary to understand and control the dynamics of hot electron and hole relaxation, cooling, charge transport, and interfacial charge transfer of the photogenerated carriers with femtosecond (fs) to ns time resolution. At NREL, we have been studying these fundamental dynamics in various bulk and nanoscale semiconductors (quantum dots (QDs), quantum rods/wires, and quantum wells) for many years using fs transient absorption, photoluminescence, and THz spectroscopy. Recently, we predicted that the generation of more than one electron-hole pair (which exist as excitons in QDs) per absorbed photon would be an efficient process in QDs . This prediction has been confirmed over the past several years in several classes of QDs by several research groups. In our laboratory we have observed very efficient and ultrafast multiple exciton generation (MEG) from absorbed single high energy photons in Group IV-VI semiconductor QDs and recently in Si QDs. Efficient MEG has the potential to greatly enhance the conversion efficiency of solar cells that incorporate QDs for both solar electricity and solar fuel (i.e. H_2) production. Selected aspects of this work will be summarized and recent advances will be discussed; a unique quantum mechanical model to explain efficient and ultrafast MEG based on the coherent superposition of multiple excitonic states (in collaboration with Al. L. Efros and A. Shabaev at NRL) will also be discussed. Various possible configurations for quantum dot solar cells that could produce ultrahigh conversion efficiencies for the production of electricity and solar fuels (e.g. H_2 from H_2O) will be presented, along with progress in developing such new types of solar cells. Finally, we have also predicted an analogous MEG effect in molecules (called singlet fission) that could be used in molecular chromophore-sensitized nanocrystalline TiO_2 solar cells and new preliminary evidence for this effect will also be presented.