Multiple Exciton Generation in Semiconductor Quantum Dots and Rods, Arrays, Buried Junctions, and Novel Molecules: Applications to Future Generation Solar Photon Conversion to Photovoltaics and Solar Fuels

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In order to utilize solar power for the production of solar electricity and solar fuels on a global 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 attain high conversion efficiencies above the well-known Shockley-Queisser thermodynamic limit of 32% is to utilize the unique properties of quantum dot/rod (QD/QR) 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. These fundamental dynamics in various bulk and nanoscale semiconductors have been studied for many years using transient absorption, photoluminescence, photocurrent, and THz spectroscopy with fs to ns time resolution The prediction that the generation of more than one electron-hole pair (which exist as excitons in size-quantized nanostructures) per absorbed photon would be an efficient process in QDs and QRs has been confirmed over the past several years in different classes of materials and their architectures. Very efficient and ultrafast multiple exciton generation (MEG), also called Carrier Multiplication (CM), from absorbed single high energy photons has been reported in Group IV-VI and Group IV semiconductors and associated solar photon conversion devices for solar electricity and solar fuels (e.g. H_2) production. Selected aspects of this work will be summarized and recent advances will be discussed. Finally, the analogous MEG effect in molecules (called singlet fission) and its use in molecular-based solar cells will also be discussed.