Multiple Exciton Generation from Single Photons in Semiconductor Nanocrystals, Quantum Dot Solar Cells, and via Singlet Fission in Molecular Chromophores: Applications to Next Generation Solar Photon Conversion to PV and Solar Fuels

Arthur. J. Nozik

National Renewable Energy Laboratory, Golden, CO 80401, USA
and
Department of Chemistry and Biochemistry, University of Colorado, Boulder 80309

Abstract

Semiconductor quantum dots (QDs), quantum rods (QRs) and unique molecular chromophores that undergo singlet fission (SF) can modify the relaxation pathways of photoexcited states to produce efficient multiple exciton generation from single photons . Efficient multiple exciton generation (MEG) has been observed in PbSe, PbS, PbTe, and Si nanocrystals and efficient SF has been observed in molecules that satisfy specific requirements for their excited state energy levels. We have studied MEG in close-packed QD arrays where the QDs are electronically coupled in the films and thus exhibit good transport while still maintaining quantization and MEG. Simple, all-inorganic QD solar cells have been developed that produce large short-circuit photocurrents and respectable power conversion efficiencies via both nanocrystalline Schottky junctions and nanocrystalline p-n junctions. These solar cells also show for the first time quantum yields (QYs) for photocurrent that exceed 100% in the photon enery regions of the solar spectrum where MEG is possible; the photocurrent MEG QYs as a function of photon energy match those determined via time-resolved spectroscopy and settles controversies concerning MEG. We have also observed very efficient SF in thin films of molecular crystals of 1,3 diphenylisobenzofuran with quantum yields of 200% at the optimum SF threshold of 2Eg (HOMO-LUMO for S_0-S_1)), reflecting the creation of two excited triplet states from the first excited singlet state. Various possible configurations for novel solar cells based on MEG in QDs and SF in molecules that could produce high conversion efficiencies will be presented, along with progress in developing such new types of solar cells. Recent analyses of the dramatic effects of MEG or SF combined with solar concentration on the conversion efficiency of solar cells will also be discussed. The properties required for nanocrystals and SF molecules to achieve the high solar conversion efficiencies predicted by theory will be presented.