MEG-based solar cells displaying EQE greater than 100%

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Photovoltaic (PV) cells are an attractive platform to provide a clean and renewable source of electricity for the growing and energy-seeking world. However, one current drawback is that low cost PV technologies are relatively inefficient and thus the technology as a whole has trouble competing with conventional "dirtier" forms of energy such as coal. Traditional semiconductors used in photovoltaic devices produce one electron from each absorbed photon. The electron is then extracted from the semiconductor at a fixed potential where the electron has lost any energy exceeding the bandgap of the semiconductor as heat. Heat loss accounts for nearly 50% of the incident insolation, and in addition to other forms of loss, a conventional silicon solar cell is thus limited in conversion efficiency to 31%.

On the other hand, new materials such as quantum dots, nanorods, carbon nanotubes and graphene can more efficiently convert high-energy photons into multiple electron-hole pairs through a process termed multiple exciton generation (MEG). This process has been shown to be more efficient in highly confined quantum dots than other forms of carrier multiplication (such as impact ionization) in bulk materials. Photovoltaic devices can benefit greatly from MEG by producing increased photocurrent from the multiple electrons and thus allowing a single junction solar cell to yield a theoretical maximum efficiency as high as 44% compared to 33% for bulk semiconductors.

MEG was first predicted to be more efficient in highly confined quantum dots than in bulk materials in 2001 by Nozik. In 2004, Schaller and Klimov demonstrated the effect using transient absorption on colloidal PbSe quantum dots. In this experiment, a high photon fluence was first used to characterize the biexciton dynamics. Then the sample was subjected to higher energy photons whilst the fluence was drastically reduced such that each QD only absorbed at most one high-energy photon. The same biexciton characteristics were seen in the high photon energy as in the high fluence experiments implying that multiple excitons were still present in the QDs although they only had absorbed one photon in the high-energy case. A slew of other materials (PbS, PbTe, CdSe, InAs, InP, Si, carbon nanotubes, graphene etc.) and optical techniques were then examined/employed in the ensuing years and differences in quantum yields among groups led to a controversy on the actual efficiency of the process and how useful it could actually be for photovoltaic applications.

In this talk, we will present recent experimental results where PbSe and PbS quantum dots are very carefully examined and compared to bulk semiconductors [Beard, MC et al. ***Nano Lett.*** 2010]. We will present how best to compare the quantum yields between different materials with differing bandgaps. We have recently reported findings from incorporating PbSe quantum dots (QDs) into semiconducting arrays that make up the absorber layer in prototype solar cells [Semonin, OE et al. ***Science***. 2011]. In these devices, MEG (and the efficiency associated with it) is confirmed by demonstrating the first solar cell with external quantum efficiency (EQE) exceeding 100% for solar relevant photon energies. We will present the evolution of the device structure from the first devices utilizing a PbSe QD array which were Schottky cell designs to the heterojunction devices used here. The advancement for these devices also involves attention to the coupling strategy employed, which will be discussed. With these recent developments, the EQE in our device reaches a maximum value of 114% at 380 nm and we have employed an optical model to determine that the PbSe QD layer produces (on average) as many as 1.3 electrons per photon for this spectral region. These findings are compared to ultrafast time resolved measurements of carrier quantum yields where we find reasonable agreement despite a 15% intrinsic loss in the transport of carriers in the solar cell. We will also discuss future directions for materials designs that increase the quantum yield through more efficient MEG.