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Engineered semiconductor nanocrystals with enhanced carrier multiplication yields

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Carrier multiplication (CM) is a process whereby absorption of a single photon results in multiple electron-hole pairs (excitons). This process could benefit a number of solar-energy conversion technologies, most notably photocatalysis and photovoltaics. This presentation overviews recent progress in understanding the CM process in semiconductor nanocrystals, motivated by an outstanding challenge in this field - the lack of capability to predict the CM performance of nanocrystals based on their known photophysical properties or documented parameters of parental bulk solids. Here, we present a possible solution to this problem by showing that, using biexciton Auger lifetimes and intraband relaxation rates inferred from ultrafast spectroscopic studies, we can rationalize relative changes in CM yields as a function of nanocrystal composition, size and shape. Further, guided by this model, we demonstrate a two-fold enhancement in multiexciton yields in PbSe nanorods vs. quantum dots attributed to enhanced Coulomb interactions. We also explore the control of competing intra-band cooling for increasing multiexciton production. Specifically, we design a new type of hetero-structured PbSe/CdSe quantum dots with reduced rates of intra-band relaxation and demonstrate a four-fold boost in the multiexciton yield. These studies provide useful guidelines for future efforts to achieve the ultimate, energy-conservation-defined CM efficiencies.