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Effects of Band Structure on the Electronic and Optical Properties of Semiconductor Nanocrystals: Lead Selenide vs. Cadmium Selenide[1]

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PbSe and CdSe nanocrystals (NCs) can be synthesized with high monodispersity, have a size-tunable band gap, and can exhibit near unity photoluminescence quantum yields. These two materials have significant differences with respect to bulk band gap, crystal structure, Bohr radius, and carrier effective masses that result in very distinct energy structures. Here we perform a side-by-side comparison of the optical and electronic properties of these two materials in both the single and multiexciton regimes for NCs of comparable sizes. Femtosecond transient absorption (TA) spectroscopy is used to study inter- and intraband relaxation of photo-generated carriers including various types of Auger-relaxation processes. We have discovered that for PbSe NCs, photo-generated single excitons with sufficient energy in excess of the band gap are able to relax by producing multiple excitons (carrier multiplication). In carrier multiplication, intraband excess energy is transferred to a valence band electron that is excited into the conduction band, resulting in the formation of two or more excitons per initially photo-excited exciton [Phys. Rev. Lett. 2004, v.92, 186601/1-4]. We have found that this effect of multiexciton generation, which has never been found to occur with significant efficiency in bulk semiconductors, can occur with up to 100% efficiency in PbSe NCs depending upon the absorbed photon energy and occurs at wavelengths that are relevant to solar energy conversion. This process, which is an enabler of Generation III solar cells, has the potential to considerably increase the power conversion efficiency of NC-based photovoltaics. Pump-power dependent TA studies performed with a probe pulse tuned to near the photoluminescence maximum have revealed that both PbSe NCs and CdSe NCs can exhibit optical gain and, when incorporated into high optical quality sol-gel waveguides, are capable of producing size-tunable amplified spontaneous emission [J. Phys. Chem. B 2003, v.107, 13765-8]. Similar TA studies performed with a probe pulse tuned to the band-edge absorption feature reveal that differences in crystal structure cause the two materials to have significantly different exciton degeneracy, which directly results in different thresholds for gain.

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