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Learning the Rules of Extreme Quantum Confinement: Ultrafast Photophysics of Semiconductor Nanocrystals

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Zero-dimensional (0D) semiconductor nanocrystals (NCs) allow the realization of new types of strongly interacting multi-exciton states that do not occur in bulk semiconductors. Using sub-10 nm colloidal nanoparticles one can generate states, in which several excitons occupy a volume comparable to or smaller than the volume of a bulk exciton. Such "squeezed" exciton states are characterized by greatly enhanced mutiparticle interactions resulting from a forced overlap of electronic wavefunctions and a reduced dielectric screening. In this paper, we utilize various ultrafast optical technique in order to study spectroscopic and dynamical signatures of multiexciton states in size- and shape-controlled CdSe and PbSe NCs. Specifically, by using a series of elongated CdSe nanoparticles (quantum rods), we investigate the effect of the 0D-to-1D transition on the efficiency of multiparticle Auger recombination. Further, by applying a femtosecond photoluminescence up-conversion technique, we detect the emission from neutral and charged biexcitons and directly measure their interaction energy. Finally, we use PbSe NCs to study the effect of carrier muliplication, in which relaxation of a single, high-energy exciton produces multiple excitons. In addition to providing new insights into the physics of exciton-exciton interactions in the regime of extreme quantum confinement, our studies have a direct relevance to the number of emerging applications of NCs in such areas as optical amplification and lasing, nonlinear-optical switching, and photovoltaics.