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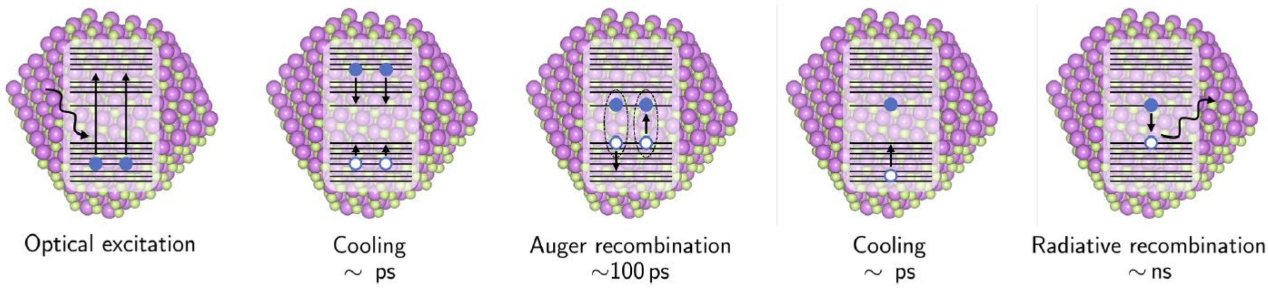

SIMULATING EXCITONS AND MULTIEXCITONS UNDER CONFINEMENT

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Semiconductor nanocrystals (NCs) are small bright light-emitting particles containing thousands of atoms and tens of thousands of valence electrons with discrete spectra at low excitation energies similar to molecules that converge to the continuum bulk limit at higher energies. Due to the quantum confinement effect, a spatial enclosure of the charge carriers (electrons, holes, excitons, etc.) within the NC, the electronic and optical properties can be tuned by simply changing the size and shape of the NCs, in addition to changing the material composition. Significant progress in developing a broad range of applications-based NC devices utilizing the quantum confinement in solar energy conversion, optoelectronic devices, molecular and cellular imaging, and ultrasensitive detection, has been hampered by the lack of a deeper understanding of the carrier dynamics under confinement.

The description of carrier dynamics in spatially confined semiconductor NCs with enhanced electron-hole and exciton-phonon interactions, is a great challenge for modern computational science. Computational methods developed for molecules are limited to very small clusters while methods for bulk systems with periodic boundary conditions are not suitable due to the lack of translational symmetry in NCs. In this talk I will review our efforts in developing atomistic approaches to describe the electronic and optical properties of semiconductor NCs and focus on the two main nonradiative loss processes: Exciton cooling and Auger recombination (see figure). I will show that electron-hole and exciton-phonon correlations are key in accurately delineating the role of size, shape, and material composition on the resulting carrier dynamics, such as the universal volume scaling of Auger recombination rates and the ultrafast, multiphonon relaxation dynamics of carriers under confinement. I will further discuss the role of size, shape, and material composition for II-VI and III-V semiconductor NCs.