Investigation of electron-hole interaction in nanoparticles using explicitly correlated wavefunction based methods

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Electron-hole pairs or excitons are generated by electronic excitation from ground to excited electronic state. Exciton dissociation and generation of free charge carriers is central for light-harvesting applications of photoactive nanoparticles. This talk will focus on computational investigation of shape-based and heterojunction-based control of electron-hole binding and recombination in quantum dots, rods, and wires. Results from a multi-faceted investigation on CdSe, CdSe/ZnS and InGaN/GaN nanoparticles using a variety of metrics including exciton binding energy, electron-hole recombination probability, electron-hole separation distance, and electron-hole pair density will be presented. These quantities were computed by solving the electron-hole Schrodinger equation using the explicitly correlated configuration interaction (XCCI) method. The XCCI method is a variational method that uses a correlated electron-hole wavefunction that depends explicitly on the electron-hole interparticle distance. Investigation of optical properties of large finite-sized cluster is computationally challenging because of the large number of electrons in the system and the absence of translational symmetry. In the present work, we show that the pseudopotential+XCCI method provide a computationally efficient route for investigating electron-hole correlation in quantum dots that are computationally prohibitive to be treated using pseudopotential+CI method. The talk will focus on the theoretical development of the XCCI method, techniques for efficient implementation, and comparison with other quasiparticle-based methods including Bethe-Salpeter equation, electron-hole variational Monte Carlo, and configuration interaction method.