Effect of the Crystal Structure on Optical Response of HgSe Quantum Dots

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Mercury selenide (HgSe) Quantum Dots (QDs) have recently attracted interest as a photodetector material due to its facile synthesis, cost-effectiveness, and exceptionally broad spectrum of tunable spectral coverage spanning from near-infrared to visible light. Such tunable light response property holds promise for both photovoltaic and optoelectronic applications. However, a comprehensive understanding of the impact of both crystal morphology and surface chemsitry on optical transitions is imperative for the practical application of HgSe. Targeting this challenge, we employ density functional theory (DFT) and time dependent DFT (TDDFT) to investigate the effect of the wurtzite versus zincblende crystal structures of stoichiometric HgSe QDs of 1nm to 2 nm in sizes on their ground and excited properties. All QD structures exhibit a minimally perturbed crystal lattice symmetry. As QD's size increases, absorption spectra consistently exhibit a redshift in energies of the brightest transitions, aligning with the confinement effect. However, wurtzite structures display more optically inactive transitions at the lowest energy spectral range, potentially arising from surface-localized 'trap' states, compared to zincblende QDs, except for the smallest ~1 nm QDs. The presence of a manifold of low-energy optically inactive states in wurtize QDs suggests competition between radiative emission to ground-state and non-radiative relaxation, which is expected to significantly diminish their emission efficiency, compared to zincblende QDs. Our calculations offer atomistic insights into structure-property relationships of QDs important for controlling the optical properties of HgSe QDs through structural engineering and provide a basis for the rational design of nanostructure for future technologies.