Developing an *Ab Initio* Toolbox to Study Trap States in Quantum Dots

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The performance of semiconductor nanocrystal quantum dots is limited by nonradiative charge recombination arising from a class of poorly understood electronic states known as trap states. Due to their large but finite size, quantum dots remain outside the range of traditional *ab initio* electronic structure methods, with even the smallest realistically-sized quantum dots being infeasible with anything other than ground-state density functional theory (DFT). Here, we present a series of approaches for circumventing this bottleneck to gain insight into quantum dot trapping. First, we present a procedure to identify information about trap states from ground-state DFT calculations, and leverage calculations over a large dataset to gain insight into the trap states in InP and GaP QDs. Second, we discuss a framework for computing X-ray photoelectron spectra of QDs through the ΔSCF formulism. Finally, we introduce a method, BYND, for computing entire excitation spectra of systems of QD size and beyond by combining real-time TD-DFT and the small-matrix approximation.