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Exciton-Assisted Relaxation of Hot Carriers in Semiconductor Quantum Dots

Hadassah B. Griffin, Andrei B. Kryjevski, Svetlana V. Kilina and Dmitri S. Kilin

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Abstract

The work provides computational arguments in support of excitonic approach for the treatment of the photo-induced processes in semiconductor quantum dots. The non-radiative relaxation, non-radiative recombination, and photo-luminescence quantum yield are computed for a range of atomistic models of semiconductor quantum dots (QDs) in the quantum confinement regime. The excitonic (EX) approach is compared to independent orbital approximation (IOA) approach. Both approaches address dissipation of the electronic energy from electronic degrees of freedom to thermal vibrations of the lattice. The difference of two approaches appears in treatment of energies of electronic states and in a way how the electron-phonon interaction is taken into account. IOA approach uses energies of Kohn-Sham orbitals and on the fly non-adiabatic couplings. [1-3] EX approach uses Bethe-Salpeter equation (BSE) for energies. [4-6] The excitonic wavefunctions from BSE is used to construct a linear transformation matrix that transforms IOA-based non-adiabatic couplings into an excitonic basis. Both approaches are compared in application to ultrasmall 1 nm diameter Si QD. Results include an evidence that hot excitons relax sooner in the excitonic picture than in the IOA picture. The observed effect is rationalized via smaller subgaps and different available relaxation pathways in the excitonic picture. The most surprising result is found for the simulated emission spectrum. The spectrum in the excitonic picture demonstrates intensity in several 5 orders of magnitude higher than in the IOA picture. This observation is related to formation of a bright exciton in the lowest excitation of the ultra-small Si QDs. Obtained evidence favors excitonic approach and promises a reliable interpretation and prediction of time-dependent observables in a range of semiconductor quantum dots of different composition, sizes, and surface environment. [7] Most intriguing results are expected for QDs representing interface between PbSe and CdSe. [8]

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