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Analysis of phonon-assisted dynamics of photoexcitations in semiconductor quantum dots

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The time evolutions of photoexcitations in PbSe and CdSe semiconductor quantum dots (QD) are calculated by (i) the ab initio non-adiabatic surface-hopping method [1], (ii) propagating a system of Pauli master equations for photoactivated states at the edge of conduction and valence bands of a QD [2], and (iii) Quantized Hamiltonian Dynamics Method. In the latter, the time evolution of the photoexcitation and the relevant population inversion are formulated in terms of Heisenberg equations of motion for conventional spin and phonon operators [3]. Depending on character of vibrational dynamics, electronic excitation exhibits either oscillatory or overdamped behavior. We also calculate the photoluminescence (PL) of an ensemble and of a single quantum dot. The PL of an ensemble of QDs shows gradual change in energy and intensity. For a single QD, intensity of PL experiences fluctuations. Our results provide physical insights regarding phonon-assisted relaxation and spectroscopy in quantum dots.

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