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Phonon-Induced Dephasing of Excitons in Semiconductor Quantum Dots: Multiple Exciton Generation, Fission, and Luminescence ANGELINE MADRID, Univ of Washington, HYEON-DEUK KIM, Kyoto University, BRADLEY HABENICHT, OLEG PREZHDO, Univ of Washington — Phonon-induced dephasing processes that govern optical line widths, multiple exciton (ME) generation (MEG), and ME fission (MEF) in semiconductor quantum dots (QDs) are investigated by ab initio molecular dynamics simulation. Using Si QDs as an example, we propose that MEF occurs by phonon-induced dephasing and, for the first time, estimate its time scale to be 100 fs. In contrast, luminescence and MEG dephasing times are all sub-10 fs. Generally, dephasing is faster for higher-energy and higher-order excitons and increased temperatures. MEF is slow because it is facilitated only by low-frequency acoustic modes. Luminescence and MEG couple to both acoustic and optical modes of the QD, as well as ligand vibrations. The detailed atomistic simulation of the dephasing processes advances understanding of exciton dynamics in QDs and other nanoscale materials.

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