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The shape and symmetry dependence of excitonic radiative decay in CdSe nanocrystals QINGZHONG ZHAO, ALBERTO FRANCESCHETTI, PETER A. GRAF, WESLEY B. JONES, KWINSEON KIM, National Renewable Energy Laboratory, LIN-WANG WANG, Lawrence Berkeley National Laboratory
— For the last decade, the exciton recombination dynamics of nanocrytal quantum dots (NQDs) through radiative decay have been extensively studied by experimental and theoretical methods, because some NQDs, like CdSe and CdS/ZnS, show near unity quantum yield. Using atomistic, semiempirical pseudopotential calculations, we investigate the radiative decay of band-edge excitons in CdSe nanocrystals with perfect and imperfect shapes. While the lifetimes of bright excitons are in the nanosecond range and not sensitive to size and shape, we find that the radiative lifetimes of the ground state dark excitons are highly dependent on the surface shape and symmetry. The introduction of one [100] or [101] facet can drastically reduce the dark exciton lifetime from milliseconds to microseconds, and such faceting is observed by STEM. This provides an alternative to the explanation by spin-flip assisted or surface-assisted recombination mechanisms of the observed microsecond dark exciton lifetime. Our results highlight the importance of QD surface shape and broken symmetry in exciton dynamics.

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