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Radiative lifetime of excitons in CdSe quantum dots M. CALIFANO, A. FRANCESCHETTI, A. ZUNGER, National Renewable Energy Laboratory, Golden, CO 80401 — Recent experimental measurements have shown that the low-temperature (T $\sim$ 2K) recombination lifetime of excitons in CdSe nanocrystal quantum dots is relatively short, of the order of 10^{-6} s for quantum dots in the 2-4 nm size range. These results are surprising, since the lowest excitonic state of CdSe quantum dots is optically "dark," and the next, "bright" state is several meV higher in energy, so it is not thermally populated at low temperature. Using a semi-empirical pseudopotential approach, we have investigated the exciton radiative lifetime of CdSe quantum dots as a function of size and temperature. We find that indeed, in the case of fully passivated CdSe nanocrystals the low-temperature lifetime is at least three orders of magnitude longer than the experimental value. However, we also find that the presence of surface states, such as dangling-bond states, mixes the dark and bright exciton states, dramatically reducing the dark exciton lifetime, and bringing it in agreement with experimental data. We conclude that surface states are the controlling factor of dark-exciton lifetimes in colloidal CdSe dots.

Alberto Franceschetti
National Renewable Energy Laboratory

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