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The Effect of Surface Ligands on Charge Carrier Dynamics in Semiconductor Nanoclusters SVETLANA KILINA, Los Alamos National Laboratory, SERGEI IVANOV, Sandia National Laboratory, OLEG PREZHDO, University of Washington, SERGEI TRETIAK, Los Alamos National Laboratory —
Using Density Functional Theory we investigate the impact of passivating ligands on morphology, electronic structure, and radiative and nonradiative dynamics in CdSe quantum dots (QDs). We consider the $Cd_{33}Se_{33}$ cluster – within a range of the smallest synthesized CdSe QDs, passivated by either amine ore phosphine oxide, modeling the common ligands used for the QD surface passivation. Our calculations show surface reorganization of both the bare and ligated clusters. The surface-ligand interactions leads to charge redistribution and polarization on the surface and result in the development of hybridized states, with the electronic density distributed over the CdSe cluster and the ligands. Neither the ligand nor hybridized molecular orbitals appear as trap states near the band gap of the QD. Instead, being optically dark, hybridized states open new relaxation channels for high-energy photoexcitations. Hybridized states are denser at the edge of the QD conduction band of the cluster ligated with phosphine oxide than that with amines. Such difference in electronic structure impacts electron-phonon interactions and non-adiabatic couplings and leads to faster electron relaxation rate in QDs passivated by phosphine oxide than by amine.

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