Effect of surface defects on electronic and optical properties of silicon quantum dots

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We investigated the effect of a surface defects, such as losing ligands and external charge, on the electronic structure and the energy transfer between methyl passivated silicon quantum dots (QDs). We have simulated different silicon quantum dots that consists of either 29, 35 or 66 silicon atoms as the QD model with a diameter ranging from 1.3 to 1.6 nm to perform DFT and linear response TDDFT calculations. Our calculations show that the surface defects introduce mid-gap states that are weakly optically active resulting in a lowest energy red shifted absorption band. We then have explored the Forster resonance energy transfer (FRET) rates between QDs of different sizes, different surface passivation, and at different distances. Our results show that introducing charge on the QD and removal of ligands from the QD surface increases the rate of energy transfer compared to ideal QDs The higher rates of energy transfer between QDs due to surface defects when packed closely facilitate occupation of the mid-gap trap states leaving the room for direct emission from higher-energy bright states. Such ultrafast energy transfer between QDs with imperfect surface might explain the nature of on-time increase in quantum blinking of closely packed Si QDs observed experimentally.

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