Atomistic theory of multi-million atom crystal phase quantum dots

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Abstract

We demonstrate an atomistic theory for calculation of excitonic properties of crystal phase quantum dots formed by a zincblende section embedded in a wurtzite nanowire. We utilize an empirical tight-binding model to account for the crystal field splitting, increased the band-gap and valence band offset between the wurtzite and zincblende segments. The single particle computation is than followed by a configuration interaction method for many-body states calculation to obtain the excitonic optical spectra. We calculate spectral properties of nanosystems containing up to 5 million atoms.

We show that the single particle structure of the electron states confined in the zincblende segment closely resembles that of self-assembled or nanowire quantum dots. On the contrary the structure of the single particle hole states is very different from the electron states. They are delocalized over the wurtzite part of the nanowire and the energy spacing between subsequent hole states are small due to reduced confinement in wurtzite sections. Moreover the hole states localized in the bottom and upper wurtzite sections of the nanowire are coupled via the zincblende barrier to form anti-bonding and bonding states of quasi-molecular-like character, that are delocalized over the entire system. Next, we calculate the optical spectra and find that the spectra strongly depend on the length of the zincblende section and on the lengths of the surrounding wurtzite matrix sections. We find that for larger diameter systems the energy spacing is reduced such that a simplified single particle picture reaches the limits of its validity and must be replaced by a full many-body approach that accounts for the configuration mixing. Large diameter crystal phase quantum dots constitute therefore a complicated, highly-correlated system.