Photoexcitations in arrays of semiconductor QDs: DFT computation

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We use Density Functional Theory (DFT) combined with the many body perturbation theory to calculate optical and spatially non-uniform field response in several hydrogen-passivated silicon nanosystems. Specifically, $Si_{29}H_{36}$ quantum dots (QDs) with crystalline and amorphous structures, the quasi one dimensional (1-D) arrays constructed from these QDs, as well as crystalline and amorphous Si-nanowires have been investigated with the main focus on their optical and electronic properties as a function of their morphology and structural disorder. We observe strong dependence of absorption spectra on the array's structure, such as QD-QD distances and mutual orientation of QDs. Organization of QDs into 1-D arrays leads to overall brightening of the lowest transitions. In 1-D QD chains, we compute response to a spatially non-uniform time dependent electric field that vanishes on every other QD. Current density induced by such electric field configurations serves as a characteristic of QD coupling in the arrays and, in particular, of the inter QD exciton transport. We observe rapid decay of the QD-QD coupling with the increase of inter QD separation.

Also, we use the same approach to calculate impact ionization (inverse Auger) bi-exciton generation rates in $Si_{29}H_{36}$ QDs and 1-D arrays. We find 1-D QD arrays to possess more efficient bi-exciton generation compared to the isolated QDs. This is in qualitative agreement with the recent experimental results on silicon QD assemblies dispersed in silicon dioxide. Bi-exciton generation in amorphous QD arrays is significantly enhanced compared to their crystalline counterparts.