Enhanced Multiple Exciton Generation in Amorphous Silicon Nanowires and Nanofilms: DFT-based Computation

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Abstract

Multiple exciton generation (MEG) in nm-sized hydrogen-passivated silicon nanowires (NWs), and quasi two-dimensional nanofilms depends strongly on the degree of the core structural disorder as shown by the many-body perturbation theory calculations based on the DFT simulations for which we use the HSE06 exchange-correlation functional. In perturbation theory, we work to the 2nd order in the electron-photon coupling and in the (approximate) RPA-screened Coulomb interaction. We also include the effect of excitons for which we solve the Bethe-Salpeter equation. To describe MEG we calculate exciton-to-biexciton as well as biexciton-to-exciton rates and quantum efficiency (QE), the average number of excitons created by a single absorbed photon. We consider 3D arrays of $Si_{29}H_{36}$ quantum dots, NWs, and quasi 2D silicon nanofilms, all with both crystalline and amorphous core structures. Also, we study chiral single-wall carbon nanotubes (SWNTs) (6,2) and (10,5). Efficient MEG QE of 1.3 up to 1.8 at the photon energy of about $3E_g$, where E_g is the gap, is predicted in these nanoparticles except for the crystalline NW and film where $QE \simeq 1$. In amorphous NWs and SWCNTs the exciton-to-biexciton rates reach $10^{14} - 10^{15}$ $1/s$ above the energy threshold. The biexciton-to-exciton recombination rates are suppressed by a factor of at least 10. MEG in the amorphous nanoparticles is enhanced by the electron localization due to structural disorder. The exciton effects significantly red-shift QE vs. photon energy curves. Nanometer-sized amorphous silicon NWs and films are predicted to have effective MEG within the solar spectrum range. Also, we find efficient MEG in the SWNTs within the solar spectrum range but its strength varies strongly with the excitation energy, which is due to highly non-uniform densities of electronic states.