Elucidating the role of non-radiative processes in charge transfer of model core-shell Si:SiO_2 nanoparticles

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Crystalline silicon is the most commonly used material in solar panels, but it is limited by its cost, band gap and the fact that it is an indirect absorber of light. The third generation approach of using inexpensive, non-toxic materials with layers that absorb different parts of the solar spectrum has the potential to dramatically increase efficiencies and lower consumer costs. Core-shell Si:SiO_2 nanoparticles are ideally suited for this application, and have been synthesized by a variety of groups in an array of sizes allowing for absorption in a wide range. A theoretical investigation of fundamental processes in these systems can guide synthetic work, and potentially lead to improved devices. Calculations on a model core-shell interface with the formula $Si_{264}O_{160}$ which features a silicon layer sandwiched between two SiO_2 layers were performed using the Vienna Ab Initio Software package (VASP)[1]. The model obeys periodic boundary conditions in three dimensions. The PBE functional in the basis of plane waves was used along with pseudopotentials to simulate electronic structure, and nuclear motion was considered using ab initio molecular dynamics as implemented in VASP. The density of states, absorption spectrum, partial charge densities, and radiative recombination lifetimes has been calculated. This interface shows quantum confinement in the pure Si part of the model[2], and exhibits behavior similar to a 1D particle in a box. The role of annealing as it affects crystallinity is explored, and the role of phonons in charge transfer and nonradiative recombination is determined. From these results, we can gain a better understanding into charge transfer processes in core-shell systems, leading to potentially better solar technology.

1 Kresse, G., and Furthmuller, J.: 'Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set', Computational Materials Science, 1996, 6, (1), pp. 15-50

2 Kilin, D.S., and Micha, D.A.: 'Surface Photovoltage at Nanostructures on Si Surfaces: Ab Initio Results', Journal of Physical Chemistry C, 2009, 113, (9), pp. 3530-3542