Abstract for the 2016 Sanibel Symposium

Quantum Confinement Effects on Photomobilities in Nanostructured Silicon Slabs Calculated with a Density Matrix Treatment [a]

Robert Hembree,[1] Tijo Vazhappilly,[2] and David A. Micha[3]

[1]Department of Chemistry and Chemical Theory Center, University of Minnesota, Minneapolis, Minnesota
[2] Theoretical Chemistry Section, Bhabha Atomic Research Center, Mumbai, Maharashtra, India
[3] Quantum Theory Project, Departments of Physics and of Chemistry, University of Florida, Gainesville, Florida

Density of states and light absorbance properties show drastic change when the number of layers of nanostructured silicon slabs are changed in the computational modeling of their atomic structures. This leads also to large changes in the number of conduction electrons and holes created by photoexcitation. We have developed a general computational procedure to calculate the mobility of photoexcited electrons and holes in semiconductor slabs using a combined density matrix and electronic band structure treatment.[1] This has been applied to slabs generated by periodic translation of 6x4 Si(111):H supercells with several layers, to generate slabs for varying thickness (from 4 to 12 layers) both with and without absorbed silver clusters.[2] Electronic band states were generated in the $\Gamma \rightarrow X$ direction with *ab initio* density functional theory in a plane-wave basis using PBE exchange and correlation density functionals, corrected to give accurate band gaps as obtained from HSE functional calculations. A steady state reduced density matrix including dissipative effects provides photoexcited populations of carriers in electronic bands in response to the absorption of photons in the visible and near IR ranges, and carrier currents are obtained from electronic displacements due to applied voltages. We find that the adsorption of small silver clusters to the surface of the Si(111) slab creates energy localized density of states for few layers, suggesting quantum confinement, and enhances photomobilities of electrons and holes. With increasing slab thickness this effect is reduced as new states appear in the bandgap and silver cluster states become delocalized into the silicon slab. Increasing the size of the silver cluster on the thicker slabs again displays the quantum confinement effect. This behavior is relevant to the development of photovoltaic materials where photoexcited electronic populations and photomobilities affect the efficiency of light absorption and electronic current transport.

[1] T. Vazhappilly, R. Hembree, and D. A. Micha, J. Chem. Phys., accepted to appear (2015)
[2] R. Hembree, T. Vazhappilly, and D. A. Micha, to be submitted for publication (2015)

[a] Work partly supported by the NSF and Dreyfus Foundation, and the HPC of UF