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Non-adiabatic Dissipative Photoexcitation of Silicon Nanostructured Surfaces

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We have employed *ab initio* electronic structure calculations and a reduced density matrix (RDM) theory to calculate the dissipative electron dynamics of photoexcitation in Si slabs, extending a recent treatment which gave very good agreement with experimental measurements of light absorbance and of photovoltages [1]. These were obtained from steady state solutions of the equations of motion for the RDM with dissipative rates due to electron-phonon interactions. Ab-initio molecular dynamics calculations have been performed to generate the electron-phonon coupling in Si slabs [2]. We employed density functional theory with a plane wave basis set and the PW91/GGA functional for the exchange-correlation to generate a basis set of Kohn-Sham orbitals and to construct matrix elements of physical operators. Changes in absorption and magnitude of electronic charge transfer have been investigated for slabs with dopants and adsorbates. The densities of electron and hole states created by steady photoexcitation of c-Si with and without silver adsorbates have been obtained. The role of localized surface plasmons has been investigated for large silver clusters. The convergence of optical properties of Si slabs with slab thickness has also been investigated.

1. T. Vazhappilly, D. S. Kilin, and D. A. Micha *J. Phys. Chem. C* (2012) *116*, 25525
2. D. S. Kilin and D. A. Micha, J. Phys. Chem. C (2009) *113*, 3530

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