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Density Matrix Treatment of Surface Photovoltage: Applications to Adsorbates on a Si Surface

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The time dependent reduced density matrix (TDRDM) method provides a useful tool for a rigorous description of the evolution of electronic densities and optical response at a semiconductor surface. Here we develop a TDRDM-based computational procedure to calculate the voltage between the surface and inner layers of a nanostructured semiconductor slab electronically excited by light absorption, obtaining the surface electric dipole from the steady state solution of the equation of motion for the TDRDM. Both perturbation theory [1] and a matrix partitioning technique are considered in our calculations. The density matrix is generated in a basis set of slab orbitals using density functional theory with periodic boundary conditions. Our treatment can be implemented for a wide class of photo-electronic materials relevant to solar energy capture. For example, applications to a H-terminated Si(111) surface and to clusters of Ag atoms on Si(111):H give the main features of calculated photovoltage versus incident photon energies in agreement with experimental trends [2].

1. D. S. Kilin and D. A. Micha, work submitted to Phys. Rev. Lett. and in progress (2007).

2. E. Fefer, Y. Shapira, and I. Balberg, Appl. Phys. Lett. **67** 17 (1995).

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