Ab initio study of the photocurrent at the Au/Si metal semiconductor nano-interface

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Photo induced charge transfer at the interface of two materials is a fundamental process in photovoltaic applications. In this study, a model of simplified photovoltaic cell composed of the periodic array of Au/Si nanostructure co-doped by Al and P[1] ($-Au_{19}-Al_1H_{36}P_1Si_{36}-Au_{19}-$) was considered, so that each simulation cell has two metal-to-semiconductor interfaces: upper and lower interface. Photoinduced time-dependent electric current was computed at *ab initio* level through the combination of continuity equation and time-dependent density matrix methodology. All calculations are made in the basis of Kohn-Sham orbitals computed in DFT with the PBE functional as implemented in the VASP software package under periodic boundary conditions. Non-adiabatic couplings between basis states are computed on-the-fly along the nuclear trajectory. A dissipative equation of motion for the reduced density matrix for electronic degrees of freedom is used to study the phonon-induced relaxation of hot electrons in the simulated system.[2] Charge carrier dynamics induced by selected photo-excitations show that hole relaxation in energy and in space is much faster than electron relaxation. Computed electric current shows specific spatial symmetry. Specifically, the current at upper interface has positive sign, and the current at lower interface has negative sign. Overall net charge transfer across the model is small, however local currents at Si/Au interfaces are substantial. In future, we plan to apply the same methodology (and compare it with NEGF) for estimating open circuit voltage, exploring I-V curves at non-zero bias, and establishing an *ab initio* methodology for computing efficiency of a PV cell made of a given pair of materials.

Reference

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