

Abstract for an Invited Paper
for the MAR10 Meeting of
The American Physical Society

Time-domain ab initio studies of photoinduced electron dynamics in nanoscale semiconductors

OLEG PREZHDO, University of Washington

Design of novel materials for energy harvesting and storage requires an understanding of the dynamical response on the nanometer scale. We have developed state-of-the-art non-adiabatic molecular dynamics techniques and implemented them within time-dependent density functional theory in order to model the ultrafast processes in these materials at the atomistic level and in real time. Quantum dots (QD) are quasi-zero dimensional structures with a unique combination of molecular and bulk properties. As a result, QDs exhibit new physical phenomena such as the electron-phonon relaxation bottleneck and carrier multiplication, which have the potential to greatly increase solar cell efficiencies. Photoinduced charge separation across molecular/bulk interfaces drives the dye-sensitized semiconductor solar cell. A subject of active research, it creates many challenges due to the stark differences between the quantum states of molecular and periodic systems, as well as the different sets of theories and experimental tools used by physicists and chemists. Our time-domain atomistic simulations create a detailed picture of these materials. By comparing and contrasting their properties, we provide a unifying description of quantum dynamics on the nanometer scale, resolve several highly debated issues, and generate theoretical guidelines for development of novel systems for energy harvesting and storage.

- [1] O. V. Prezhdo “Photoinduced dynamics in semiconductor quantum-dots: insights from time-domain ab initio studies”, *Acc. Chem. Res.*, available online.
- [2] O. V. Prezhdo, W. R. Duncan, V. V. Prezhdo, “Photoinduced electron dynamics at semiconductor interfaces: a time-domain ab initio prospective”, *Prog. Surf. Science*, **84**, 39 (2009).
- [3] O. V. Prezhdo, et al., “Dynamics of the photoexcited electron at the chromophore-semiconductor interface”, *Acc. Chem. Res.*, **41**, 339 (2008).
- [4] W. R. Duncan, O. V. Prezhdo, “Theoretical studies of photoinduced electron transfer in dye-sensitized TiO₂”, Review, *Ann. Rev. Phys. Chem.*, **58**, 143 (2007).
- [5] C. F. Craig, W. R. Duncan, O. V. Prezhdo “Trajectory surface hopping in the time-dependent Kohn-Sham theory for electron-nuclear dynamics”, *Phys. Rev. Lett.*, **95** 163001 (2005).