Ab Initio Nonadiabatic Molecular Dynamics of Wet-Electrons on the TiO$_2$ Surface$^1$ SEAN FISCHER, University of Washington, WALTER DUNCAN, Schrodinger Inc., OLEG PREZHDNO, University of Washington — The electron transfer (ET) dynamics of wet-electrons on a TiO$_2$ surface was investigated using state-of-the-art ab initio nonadiabatic molecular dynamics, which includes electronic evolution, phonon motions, and electron-phonon coupling. Delocalized over both water and TiO$_2$, wet-electrons are supported by a new type of state that is created at the interface due to the strong water-TiO$_2$ interaction and that cannot exist separately in either material. Our simulations indicate that the ET is sub-10 fs and driven mainly by low frequency vibrational modes. The high density of TiO$_2$ conduction band states leads to frequent crossings of the strongly coupled donor and acceptor states, which is conducive to fast ET. The average ET dynamics for the system feature essentially equal contributions from both adiabatic and nonadiabatic transfer mechanisms. Similar states are present in a number of other systems with strong interfacial coupling, including certain dye-sensitized semiconductors. The wet-electron state may also have relevance to the electrochemical photolysis of water.

$^1$Work funded by grants from NSF and ACS

Sean Fischer
University of Washington

Date submitted: 20 Nov 2009

Electronic form version 1.4