

Abstract Submitted  
for the ES2015 Workshop

Sorting Category: (Theoretical)

**Detection of trap states using pump-probe measurements and TD-DFT** GRAHAM CLENDENNING, University of Ontario Institute of Technology, STEPHANIE CHOING, University of California, Berkeley, AARON FRANCIS, North Carolina State University, MICHAEL SCHUURMAN, National Research Council, Ottawa, Canada, ROGER SOMMER, North Carolina State University, WALTER WEARE, North Carolina State University, TANJA CUK, University of California, Berkeley, ISAAC TAMBLYN, University of Ontario Institute of Technology — In artificial photosynthetic devices, light harvesting molecules drive reactions which create fuel (e.g by splitting water). For optimal fuel generation efficiency a long charge transfer excited state lifetime is necessary. In this study, we considered the experimentally synthesized vanadium(V) oxo compound VOLF which has been observed to remain in the optically excited state for 438 ps. We used linear response time dependent density functional theory (TD-DFT) to compute and interpret static UV-Vis spectra and ultrafast transient absorption. To assess effects due to finite temperature, we used first principles molecular dynamics to generate multiple starting configurations in different solvent environments. Comparing with experiment, we find good agreement with the observed spectra. Furthermore, we find that the bright state is associated with charge transfer to the vanadium from surrounding ligands. Our results suggest transitions between low lying d-states cause relaxation to the ground state to be both dipole and vibronically forbidden. This phenomenon gives rise to extended excited state lifetimes.

Prefer Oral Session  
 Prefer Poster Session

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Date submitted: April 1, 2015

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