Abstract Submitted
for the FWS19 Meeting of
The American Physical Society

Simulating Radiative and Non-Radiative Decay Pathways of Photoexcited Ruthenium Polypyridyl Complexes THOMAS CHESHIRE, Chemical Sciences Division, Lawrence Berkeley National Laboratory, PAUL GIOKAS, DAVID ZIGLER, M. KYLE BRENNAMAN, ANDREW MORAN, JOHN PAPANIKOLAS, GERALD MEYER, THOMAS MEYER, Department of Chemistry, University of North Carolina at Chapel Hill, FRANCES HOULE, Chemical Sciences Division, Lawrence Berkeley National Laboratory — Bottlenecks in the generation of photoinduced currents in dye-sensitized solar cells are not fundamentally understood due to the lack of detailed information on initial ultrafast processes in photoexcited dyes that compete with charge injection. The ultrafast photophysics that drive solar energy conversion are typically reported in terms of phenomenological lifetimes, yet competing transitions occurring on comparable timescales obscures the relationship between such time constants and fundamental rate coefficients. Knowledge of primary kinetics will reveal dye design strategies that may improve sensitivity and efficiency. We employ stochastic simulations, which are a form of kinetic Monte Carlo that produce an absolute time base, to model explicit photoexcitation and the subsequent relaxation pathways of a series of ruthenium polypyridyl chromophores. The initial work focuses on isolated dyes in solution, producing a scheme that can be extended to dyes adsorbed on metal oxide surfaces. We predict transient absorption signals for comparison to spectroscopic data from multiple studies, and find that common assumptions about the sub-picosecond photophysics such as singlet-triplet energy transfer efficiency do not correctly reproduce experimental observations.

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Date submitted: 27 Sep 2019 Electronic form version 1.4