Abstract Submitted
for the MAR17 Meeting of
The American Physical Society

Transient Spectra in TDDFT: Corrections and Correlations JOHN PARKHILL, TRIET NGUYEN, Univ of Notre Dame — We introduce an atomistic, all-electron, black-box electronic structure code to simulate transient absorption (TA) spectra and apply it to simulate pyrazole and a GFP chromophore derivative[1]. The method is an application of OSCF2, our dissipative extension of time-dependent density functional theory. We compare our simulated spectra directly with recent ultra-fast spectroscopic experiments, showing that they are usefully predicted. We also relate bleaches in the TA signal to Fermi-blocking which would be missed in a simplified model. An important ingredient in the method is the stationary-TDDFT correction scheme recently put forwards by Fischer, Govind, and Cramer which allows us to overcome a limitation of adiabatic TDDFT. We demonstrate that OSCF2 is able to predict both the energies of bleaches and induced absorptions, as well as the decay of the transient spectrum, with only the molecular structure as input. With remaining time we will discuss corrections which resolve the non-resonant behavior of driven TDDFT, and correlated corrections to mean-field dynamics.

[1]T. Nguyen J. Koh and J. Parkhill Journal of Physical Chemistry Letters, 2016

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Date submitted: 19 Sep 2016 Electronic form version 1.4