NEXMD v2.0 Software Package for Nonadiabatic Excited State Molecular Dynamics Simulations.

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Supplementary Information for

NEXMD v2.0 Software Package for Nonadiabatic Excited State Molecular Dynamics Simulations

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1. Computational and technical details for the exemplary simulations

The electronic structure was performed in the at the CIS/AM1 level of theory. The conformational sampling was generated by performing a 1 ns long ground state dynamics at 300 K using a Langevin thermostat with a friction coefficient of $20\ \mathrm{ps}^{-1}$. The time step for these dynamics was set to 0.5 fs. One thousand equally-spaced snapshots were collected at 1 ps intervals to calculate the linear absorption spectra shown in **Figure 1**. In agreement with previous convergence tests, 300 of these snapshots were randomly selected and then used as initial conditions for exited state dynamics, which in all cases started from a pure state. For TSH dynamics we use a classical time step of 0.1 fs while it was reduced to 0.05 fs for EHR and AIMC dynamics. All other input parameters for excited state dynamics were set to default values.

All simulations were run at Los Alamos National Laboratories Chicoma supercomputer. The 1 ns ground state molecular dynamics took approximately 3 days. The TSH dynamics taking into account two excited states took approximately 6 hours. The EHR dynamics taking into account two excited states took approximately 12 hours. Finally, the AIMC dynamics computation time strongly depends on the number of bifurcations generated, such that each new one increases the total computation time as adding a new EHR dynamics. We allowed a maximum number of 16 trajectories per initial condition, so AIMC are approximately one order of magnitude slower than EHR dynamics.

2. Comparison of non-adiabatic excited state methods when exciting to S3

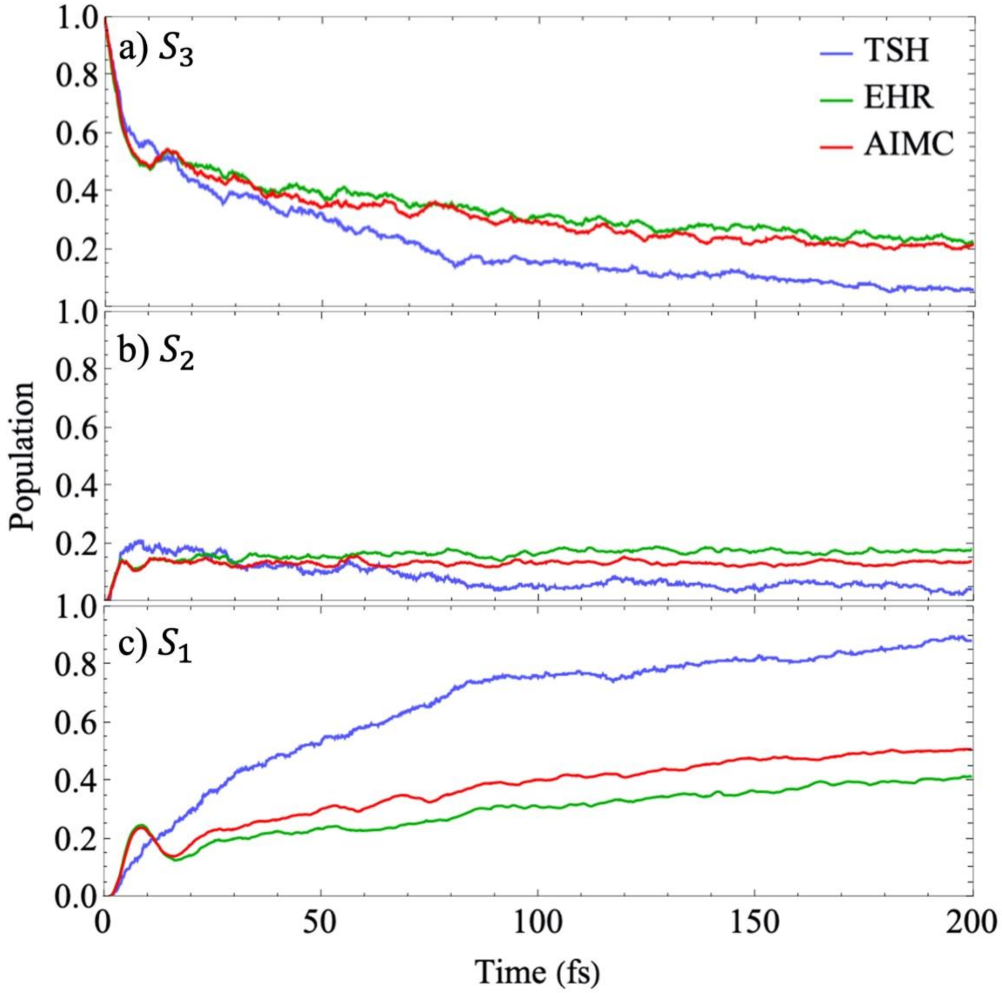

Figure S1. Evolution in time of the populations of a) S_3, b) S_2 and c) S_1 when the system is excited initially at S_3 for different nonadiabatic molecular dynamics methods.