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Nonadiabatic Dynamics Study of the B850-B800 Complex in LH2 using TeraChem Cloud

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Dynamic processes involved in excitation energy transfer (EET) are the foundation for understanding biological and artificial photosynthesis in multichromophoric systems like the LH2 complex from purple bacteria. The *ab initio* exciton model provides a scalable approach for these large systems, which would otherwise be prohibitively expensive even for excited state methods such as time-dependent density functional theory (TD-DFT). Using the point-dipole approximation, the Hamiltonian matrix elements of the *ab initio* exciton model can be expressed as simple electrostatic interactions between monomers involving energies, dipoles, and transition dipoles[1-2]. The full excitonic Hamiltonian is then diagonalized and use to run atomistic nonadiabatic dynamics with either surface hopping (SH) or *ab initio* multiple spawning (AIMS).
The *ab initio* exciton model leverages TeraChem Cloud to take advantage of a multilevel parallelization scheme where individual monomer calculations are distributed and further accelerated using graphics processing units (GPUs). TeraChem Cloud is a cloud-based platform using off-the-shelf web components, containerization technologies (i.e. Docker and Kubernetes), and the TeraChem quantum chemistry package[3-5] to provide GPU-accelerated electronic structure calculations as a service[6].
The B800-B850 assembly in LH2 consists of 27 bacteriochlorophyll-a monomers with 76 atoms each, totaling over 2,000 atoms for the entire complex. Treating each monomer with TD-DFT at the PBEh/6-31g level of theory, a single 300 fs surface hopping trajectory was completed in just over two days using a TeraChem Cloud instance deployed to 7 nodes of the XStream computational resources at Stanford University. However, more initial conditions and longer trajectories are needed to accurately describe the B800-B850 population transfer, which occurs on the 600 fs – 1 ps timescale. Increasing the capacity of our TeraChem Cloud instance with the Open Molecular Science Cloud would enable full atomistic and electronic resolution of EET processes on the picosecond timescale in LH2.

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