Time-Resolved Excitation Dynamics in a Supermolecular Light-Harvesting Complex

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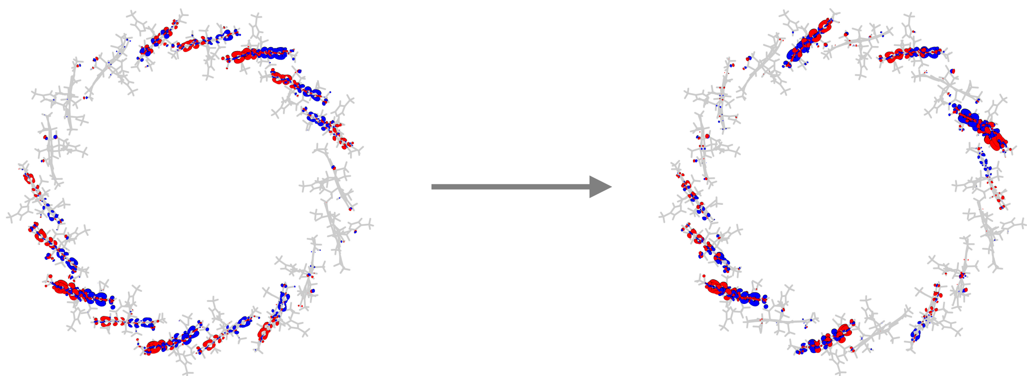
Photosynthetic light-harvesting complexes are nature's fascinating solution for efficient solar energy conversion, with antenna complexes absorbing sunlight and transferring the collected energy to a reaction center with remarkable quantum efficiency. A fundamental question is how the electronic excitation is transported through the antenna complex and what role nuclear motion at ambient temperatures plays in this process. We investigate the ultrafast excitation dynamics in the B850 antenna ring in *Rhodoblastus acidophilus* following a short laser pulse and study how nuclear vibrations interplay with the electronic excitation [1].

Using time-dependent density functional theory in real time combined with Ehrenfest molecular dynamics, we simulate, on a first-principles basis, the dynamics of the 18-Bacteriochlorophyll ring system after photoexcitation. The electronic interactions of this large biomolecular assembly (almost 2000 atoms) are explicitly included in the dynamics by treating the supermolecular complex as one entire system without relying on exciton models. We compare simulations with coupled electron-nuclear dynamics with reference calculations using frozen nuclear coordinates to isolate contributions of nuclear dynamics to the excitation-energy transfer.



Initially after the laser pulse, the excitation is delocalized over almost the whole ring. Quantum interference patters appear after about 40 fs, which leads to a transient localization of the excitation energy. On the same time scale, nuclear motion noticeably influences the excitation dynamics and the B850 ring transitions into a regime in which the excitation energy is mainly localized on segments that comprise just a few Bacteriochlorophyll molecules.

[1] T. Trepl, I. Schelter, S. Kümmel, *J. Phys. Chem. Lett.,* **2025,** *16*(42), 10891-10898.