

# Simulation of charge transfer in complex systems:

## Merging model Hamiltonian approaches with electronic structure calculations and classical molecular dynamics simulations

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2:00 pm

Room 8335



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In the last years, we have developed a computational methodology to simulate charge transfer processes in complex systems. First applications were concerned with charge transfer in DNA, which has received much attention in the last years due to its role in oxidative damage and repair in DNA, but also due to possible applications of DNA in nano-electronics. Currently, we are extending and applying the methodology to study CT in organic materials. Charge-transfer (CT) parameters are computed using a fragment orbital approach applying the approximate Density Functional method SCC-DFTB. Environmental effects are captured using a combined quantum mechanics/molecular mechanics (QM/MM) coupling scheme and dynamical effects are included by evaluating these CT parameters along extensive classical molecular dynamics (MD) simulations. Using this methodology, the time course of the hole can be followed by propagating the hole wave function using the time dependent Schrödinger equation for the Tight Binding Hamiltonian, which can also be used to compute the transmission and current through DNA nano-wires with the Landauer formalism. We apply the methodology to study the photoactivation of E. coli Photolyase which involves, after photoexcitation of the chromophore and energy transfer to FAD, a long range hole transfer along a chain of Trp residues. Since this process could not be modeled using Marcus theory with parameters computed with classical equilibrium MD simulations, we used fully coupled nonadiabatic (Ehrenfest) quantum mechanics/molecular mechanics (QM/MM) simulations. Charge transfer rates are in excellent agreement with experimental data and the simulations provide a more detailed picture of electron transfer than a classical analysis of Marcus parameters. The protein and solvent both strongly influence the localization and transport properties of a positive charge, but the directionality of the process is mainly caused by solvent polarization. The time scales of charge movement, delocalization, protein relaxation and solvent reorganization overlap and lead to nonequilibrium reaction conditions.

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