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Nonadiabatic Dynamics of Photoinduced Proton-Coupled Electron Transfer Processes in Solution[1]

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Theoretical approaches developed to elucidate the fundamental principles underlying the nonequilibrium dynamics of photoinduced proton-coupled electron transfer (PCET) processes in solution will be presented [1-3]. These processes are simulated by propagating nonadiabatic surface hopping trajectories on electron-proton vibronic surfaces that depend on the solute and solvent nuclear coordinates. The solvent is represented either as explicit solvent molecules or as a dielectric continuum. In the latter case the solvent dynamics is described in terms of two collective solvent coordinates corresponding to the energy gap coordinates associated with electron and proton transfer. Calculations on model systems reveal two distinct solvent relaxation timescales, where the faster timescale relaxation corresponds to librational motions of solvent molecules in the first solvation shell and the slower time scale corresponds to the bulk solvent dielectric response. These calculations illustrate that the charge transfer dynamics, solvent dynamics, and vibrational relaxation processes are strongly coupled. Extensions of the methodology and applications to experimentally relevant systems will also be discussed [4].

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