Photoinduced Charge Transfer Dynamics in Complex Condensed Phase

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The efficient conversion of solar energy relies heavily on ultrafast charge transfer (CT) processes, which pose computational challenges due to the complexity arising from numerous nuclear and electronic degrees of freedom. In this presentation, we showcase two recent theoretical modeling approaches for investigating photoinduced CT phenomena. First, we focus on an all-atom simulation approach applied to a prototypical organic photovoltaic (OPV) molecule dissolved in explicit tetrahydrofuran (THF) solvent. Specifically, we examine the carotenoid-porphyrin-fullerene molecular triad and employ the newly proposed Instantaneous Marcus Theory (IMT) to obtain time-resolved CT rate coefficients. Additionally, we explore nonadiabatic dynamics occurring among multiple electronic states using a semiclassical dynamical framework within the mapping basis. This approach allows us to simulate the intricate interplay between electronic and nuclear degrees of freedom at the atomistic level. Machine-learning techniques are applied to predict the CT rate constants for different molecular geometries and to predict the nonadiabatic dynamics with short-time input. Furthermore, we introduce the Multi-State Harmonic (MSH) model Hamiltonian as an effective modeling tool constructed from all-atom simulations. By employing the MSH model, we demonstrate its capability to reproduce nonadiabatic dynamics observed at the all-atom level. Our findings underscore the effectiveness of the MSH model in capturing complex nonadiabatic processes in condensed-phase environments, providing a systematic approach for understanding and optimizing solar energy conversion mechanisms.