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Light-induced nonadiabatic dynamics in molecular assemblies and nanostructures

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The combination of mixed quantum-classical dynamics with efficient electronic structure methods was developed in order to simulate the light-induced processes in complex molecules, multichromophoric aggregates and metallic nanostructures. We will demonstrate how the combination of nonadiabatic dynamics with experimental pump-probe techniques such as time-resolved photoelectron imaging (TRPEI) allows to fully resolve the mechanism of excited state relaxation through conical intersections in several prototype organic- and biomolecules. Specifically, the role of the solvent in the excited state relaxation in microsolvated and fully solvated systems will be addressed. Currently there is growing evidence that nonadiabatic relaxation processes also play a fundamental role in determining the efficiency of excitonic transfer or charge injection in multichromophoric assemblies. Since such systems are currently out of the reach of the state-of-the-art quantum chemistry a development of even more efficient quantum chemical approaches is necessary in order to describe the excited state dynamics in such assemblies. For this purpose we have recently developed long-range corrected time-dependent density functional tight binding (LC-TDDFTB) nonadiabatic dynamics and combined it with the QM/MM approach in order to simulate exciton relaxation in complex systems. The applications of the method to the investigation of the optical properties and dynamics in multichromophoric assemblies including stacked pi-conjugated organic chromophores, model molecular crystals as well as self-organized dye aggregates will be presented. Finally, we will address exciton transport dynamics coupled with the light propagation in hybrid exciton-plasmon nanostructures, which represent promising materials fort the development of novel light-harvesting systems.