Quantum Efficiency
Seminar und Colloquium

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Elementary processes of organic photovoltaics - What do we learn from a molecular-level perspective?

This talk summarizes our recent molecular-level, quantum dynamical studies of photoinduced exciton transport and exciton dissociation in functional organic polymer materials. As highlighted by recent experiments [1], these processes can be guided by quantum coherence, despite the presence of electron-phonon coupling and static and dynamic disorder. Our approach combines parametrized Hamiltonians, based on electronic structure calculations, with accurate quantum dynamics simulations using the multiconfiguration time-dependent Hartree (MCTDH) method [2]. We will specifically address (i) the dynamics of exciton transfer across a torsional defect that locally breaks the pi-conjugation in an oligo-(p-phenylene vinylene) type fragment [3], and (ii) exciton dissociation in oligothiophene-fullerene complexes [4], i.e., models for P3HT-PCBM heterojunctions, and in novel ordered donor-acceptor assemblies [5]. A unifying theoretical framework is provided by a vibronic coupling Hamiltonian in a monomer-based electron-hole representation [3] that accounts for the intrinsic coherence size of the relevant excitonic states - beyond the standard Frenkel exciton picture - and at the same time accommodates the creation of charge transfer excitons at heterointerfaces. Using these methods, a perspective will be given on the possible role of hot charge transfer excitons in the photocurrent generation.