High-Dimensional Quantum Dynamics of Functional Organic Polymer Materials: Coherence, Confinement, and Disorder

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This talk addresses quantum dynamical studies of ultrafast photo-induced energy and charge transfer in functional organic materials, complementing time-resolved spectroscopic observations [1] which underscore that the elementary transfer events in these molecular aggregate systems can be guided by quantum coherence, despite the presence of static and dynamic disorder. The intricate interplay of electronic delocalization, coherent vibronic dynamics, and trapping phenomena requires a quantum dynamical treatment that goes beyond conventional mixed quantum-classical simulations. Our approach combines first-principles parametrized lattice Hamiltonians [2], based on TDDFT and/or high-level electronic structure calculations, with accurate quantum dynamics simulations using the Multi-Configuration Time-Dependent Hartree (MCTDH) method [3] and its multi-layer (ML-MCTDH) variant [4], along with semiclassical methods [5]. The talk will specifically focus on (i) the elementary mechanism of exciton migration in oligothiophene (OT) model systems representative of the poly(3-hexyl thiophene) donor material [5-7], (ii) the creation of charge transfer excitons in stacked, regioregular $(OT)_n$ assemblies [8-9], and (iii) exciton dissociation and free carrier generation in regioregular donor-acceptor assemblies [2,9]. Special emphasis is placed on the influence of structural (dis)order and molecular packing, which can act as a determining factor in transfer efficiencies. Against this background, we will comment on the role of temporal and spatial coherence along with a consistent description of the transition to a classical-statistical regime [7].

References

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