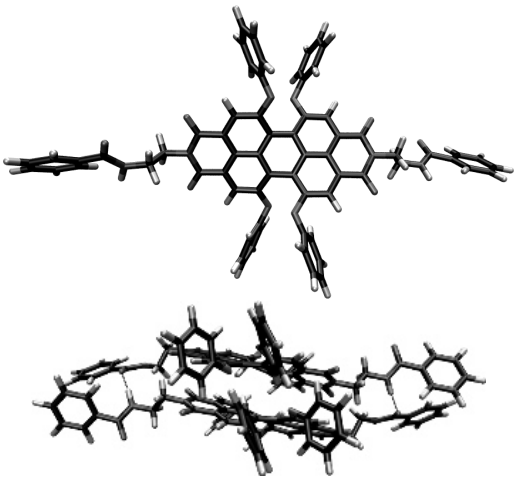
Multi-Exciton Dynamics in Molecular Aggregates

David Ambrosek, Sergey Polyutov, Yun-an Yan, Oliver Kühn
[1]*Institute of Physics, University of Rostock, D-18051 Rostock, Germany*

Excitation energy transfer in supramolecular architectures covers a broad range of dynamical regimes, depending on the electronic properties of the constituent dye building blocks, their mutual Coulomb interaction, and the coupling to vibrational degrees of freedom and to some solvent environment. In view of the size of these complexes quantitative insight can be provided only by combination of first principles simulations of parametrized models yielding spectroscopic observables.[1]

In the first part of this presentation, electronic structure theory is utilized to construct a Frenkel exciton Hamiltonian for a J-aggregate-forming perylene bisimide dye (see figure). In particular it will be shown that the optical properties are strongly dependent on the aggregate size. In accord with experimental fluorescence studies, the J-aggregate turns into an H-aggregate with decreasing number of monomers.



On the basis of this Frenkel exciton Hamiltonian quantum dynamical simulations are performed putting emphasis on the interplay between laser-driven multi-exciton generation, exciton-vibrational coupling, and exciton-exciton annihilation.

[1] V. May, O. Kühn. Charge and Energy Transfer Dynamics in Molecular Systems, 3rd Edition, Wiley-VCH, Weinheim, 2011.