Energy Transport in Natural and Synthetic Supramolecular Aggregates

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The efficient transport of excitation energy is a key process in the initial steps of photosynthesis as well as in photovoltaic applications of synthetic organic materials. In particular, the recent observation of quantum coherent effects in the ultrafast energy transport in photosynthetic light-harvesting complexes has triggered substantial attention. However, to date it is still a matter of debate whether these quantum effects are important for the efficiency of light-energy conversion, because photosynthetic complexes feature a large structural and electronic heterogeneity, which hampers the development of a molecular-scale understanding of these processes.

Here, we will discuss our recent advances in developing ultrafast techniques to study transient coherent phenomena on the level of single molecules and individual molecular aggregates. In proof-of-principle experiments, we prepared, controlled, and read-out electronic coherences on femtosecond time scales in a model system, individual terrylene molecules embedded in a polymer matrix at room temperature. Extending this technique, femtosecond energy transport was resolved within single light-harvesting antenna complexes from photosynthetic purple bacteria. We identified quantum coherent transfer that persists at least 400 fs at room temperature, and found that each individual complex features a distinct energy transfer pathway.

Finally, we will discuss how the design principles of photosynthetic complexes can be used to construct artificial supramolecular structures. Exploiting self-assembly of an organic building block, a carbonyl-bridged triarylamine, we created stable nanofibres with mono-molecular diameter that are able to transport excitation energy efficiently over up to 4 µm at ambient conditions.