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QE

Quantum Efficiency Seminar und Colloquium

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Ultrafast single-molecule detection: from small molecules to photosynthetic antenna complexes

Ultrafast energy transfer processes play a key role in the initial steps of photosynthesis as well as in photovoltaic applications of synthetic functional materials. However, the typically large heterogeneity and disordered environments of both natural and artificial macromolecules hamper a detailed nanoscale understanding of the energy transport dynamics, because conventional ultrafast techniques intrinsically average over large ensembles.
Here, we present our recent advances in combining femtosecond pulse-shaping methods with single-molecule detection schemes at room temperature. Employing this novel technique we have prepared, controlled, and read-out electronic coherences and their femtosecond decay in a model system, individual terrylene molecules embedded in a polymer matrix [1,2]. Additionally, vibrational wave-packets in single chromophores have been observed and manipulated by adapting the time and phase distribution of the laser field to the ultrafast molecular dynamics [3]. Finally, we have extended these techniques to study femtosecond coherent transport within single photosynthetic light-harvesting complexes from purple bacteria. We have identified quantum coherences between electronically coupled energy eigenstates that persist at least 400 fs at room temperature. Moreover, each individual complex features a distinct energy transfer pathway, which yields unprecedented new insights into the initial light-driven processes of photosynthesis

References:

[1] R. Hildner, D. Brinks & N. F. van Hulst, Nature Phys. **7**, 172 - 177 (2011).

[2] R. Hildner, D. Brinks, F. D. Stefani & N. F. van Hulst, Phys. Chem. Chem. Phys. **13**, 1888 - 1894 (2011).

[3] D. Brinks, F. D. Stefani, F. Kulzer, R. Hildner, T. H. Taminiau, Y. Avlasevich, K. Müllen & N. F. van Hulst, Nature **465**, 905 - 908 (2010).

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