MO 17: Excitons and Excitation Transfer (SYET 2)

Time: Thursday 14:00–15:30 Location: BEBEL HS213

Invited Talk MO 17.1 Thu 14:00 BEBEL HS213

Non-adiabatic relaxation dynamics in perylene bisimide dimers excited by femtosecond laser pulses — •Volker Engel[1], Bernd Engels[1], Mirjam Falge[1], Martin Kess[1], Stefan Lochbrunner[2], Alexander Schubert[1], Volker Settels[1], Walter Strunz[1], and Frank Würthner[3] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Emil-Fischer- Str. 42, 97074 Würzburg, Germany — [2]Institut für Physik, Universität Rostock, Universitätsplatz 3, 18055 Rostock, Germany — [3]Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

We present a model for the relaxation dynamics in perylene bisimide dimers which is based on ab-initio electronic structure and quantum dynamics calculations including effects of dissipation. The excited state dynamics proceeds via a mixing of electronic states of local Frenkel and charge-transfer characters which becomes effective upon a small distortion of the dimer geometry. In this way it is possible to explain the fast de-population of the photo-excited state as seen in femtosecond transient absorption measurements. This hints at a trapping mechanism which involves non-adiabatic and dissipative dynamics in an excited state vibronic manifold.

MO 17.2 Thu 14:30 BEBEL HS213

Hopping transport of excited states in polymeric carbon nitride photocatalysts — •Christoph Merschjann[1,2], Stefanie Tschierlei[1], Stefan Lochbrunner[1], Tobias Tyborski[2,3], Arne Thomas[4], and Thomas Schedel-Niedrig[3] — [1]Institut für Physik, Universität Rostock , D-18051 Rostock, Germany — [2]Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, D-12489 Berlin, Germany — [3]Helmholtz-Zentrum Berlin, D-14109 Berlin, Germany — [4]Technische Universität Berlin, D-10623 Berlin, Germany

The spectral and temporal development of optically excited states in polymeric carbon nitride (PCN) photocatalysts is investigated using time-resolved optical spectroscopy. By combining transient absorption results from a femtosecond pump-probe setup and transient photoluminescence using streak-camera and time-resolved single-photon counting investigations, the visible radiative emission upon UV excitation is obtained.

It is found that the decay of the emission exhibits a non-exponential behaviour over more than six decades in time (150 fs to 500 ns).

This characteristic points to a diffusive transport of localized excited states (Frenkel excitons). The underlying incoherent hopping transfer bears strong similarities to a small polaron transport. The consequences of such a comparably slow kind of transport for the catalytic activity of PCN are discussed.

MO 17.3 Thu 14:45 BEBEL HS213

Exciton coupling effects in polymeric cis-indolenine squaraine dyes — •Federico Koch[1], Sebastian Völker[2], Christoph Lambert[2], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — [2]Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

Polymeric cis-indolenine squaraine (pSQ) dye compounds display a broad absorption in the red-to-NIR region with disctinct maxima but a narrow fluorescence. The energetic shifts of the absorption maxima corresponding to the monomer can be explained by a mixture of H- and J-type alignment of chromophores [1].

We investigated the exciton coupling of pSQ in DCM (predominantly J-type alignment) and DMF (predominantly H-type alignment) by femtosecond transient absorption (TA) and coherent two-dimensional (2D) spectroscopy. In the TA studies we observed ultrafast relaxation dynamics ranging from fs to ns. The fast dynamics probably relate to an excitonic coupling. With 2D spectroscopy we aim at the direct characterization of the coupling and the corresponding interaction between the excitonic states.

[1] S. F. Völker, and C. Lambert, Chem. Mat. 2012, 24, 2541-2553

MO 17.4 Thu 15:00 BEBEL HS213

Vibronic speed-up of the excitation energy transfer in the Fenna-Matthews-Olson complex — •Peter Nalbach, Cesar A. Mujica-Martinez, and Michael Thorwart — I. Institut für Theoretische Physik, Universität Hamburg, Jungiusstr. 9, 20355 Hamburg

We show that the efficient excitation energy transfer in the Fenna-Matthews-Olson molecular aggregate under realistic physiological conditions is fueled by underdamped vibrations of the embedding proteins. For this, we present numerically exact results for the quantum dynamics of the excitons in the presence of nonadiabatic vibrational states in the Fenna-Matthews-Olson aggregate employing a environmental fluctuation spectral function derived from experiments. Assuming the prominent 180 cm^{-1} vibrational mode to be underdamped, we observe, on the one hand, besides vibrational coherent oscillations between different excitation levels of the vibration also prolonged electronic coherent oscillations between the initially excited site and its neighbours. On the other hand, however, the underdamped vibrations provide additional channels for the excitation energy transfer and by this increase the transfer speed by up to 30% . (ArXiv:1311.6363)

MO 17.5 Thu 15:15 BEBEL HS213

Temperature-dependent femtosecond time-resolved pump-probe study of exciton dynamics in (6,5) SWNTs — •Christoph Mann and Tobias Hertel — Julius-Maximilians-Universität Würzburg, Institut für Physikalische und Theoretische Chemie, Würzburg

Single wall carbon nanotubes (SWNTs) can be thought of as one-dimensional (1D) nanostructures consisting of rolled-up graphene sheets. Because of their unique electronic, mechanical and optical properties, an understanding of the exciton dominated excited-state dynamics in SWNTs is of great interest for both, better insight into fundamental photophysics of 1D systems in complex environments and for optoelectronic applications. We studied the temperature dependence of femtosecond transient absorption kinetics. In addition we carried out time-resolved and steady-state photoluminescence investigations between room temperature and 14 K. The combined information from these experiments provides new insights into the coupling of bright and dark excitonic states as well as on the temperature dependence of radiative and nonradiative processes.