

CPP 26: Molecular Electronics and Excited State Properties

Time: Tuesday 14:00–16:00

Location: ZEU 260

CPP 26.1 Tue 14:00 ZEU 260

Ultrafast Exciton Dynamics in Diindenoperylene Films — ●MARC HÄNSEL¹, VALENTINA BELOVA², KATHARINA BROCH², FRANK SCHREIBER², and PETRA TEGEDER¹ — ¹Physikalisch-Chemisches Institut, Universität Heidelberg, Germany — ²Institut für Angewandte Physik, Universität Tübingen, Germany

Diindenoperylene (DIP) is a very promising material for organic electronics. It shows a high charge-carrier mobility, forms thin films with high structural order and is resistant to most environmental influences. In a photovoltaic device it can be used as donor or acceptor material. In this work we monitored the ultrafast (sub picosecond) excited states dynamics in DIP after optical excitation using time-resolved second harmonic generation (TR-SHG). We investigated the exciton dynamics in thin DIP films on two different substrates, namely SiO₂ and Sapphire. Sapphire was used as non-interacting substrate to analyze the behavior of excitonic species in the pure DIP film. The generation dynamics of singlet excitons localized on single molecules followed by the formation of charge transfer excitons between two DIP molecules could be resolved. On the SiO₂ (native oxide layer 2nm) additional ultrafast processes were found, due to electronic coupling effects between DIP and the semiconducting substrate.

CPP 26.2 Tue 14:15 ZEU 260

Tuning nonradiative lifetimes via molecular aggregation — ●ALAN CELESTINO and ALEXANDER EISFELD — MPIPES, Dresden, Germany

We show that molecular aggregation can strongly influence the nonradiative decay (NRD) lifetime of an electronic excitation. As a demonstrative example, we consider a transition-dipole-dipole-interacting dimer whose monomers have harmonic potential energy surfaces (PESs). Depending on the position of the NRD channel (q_{nr}), we find that the NRD lifetime (τ_{nr}^{dim}) can exhibit a completely different dependence on the intermolecular-interaction strength. We observe that (i) for q_{nr} near the Franck-Condon region, τ_{nr}^{dim} increases with the interaction strength; (ii) for q_{nr} near the minimum of the monomer excited PES, the intermolecular interaction has little influence on τ_{nr}^{dim} ; (iii) for q_{nr} near the classical turning point of the monomer nuclear dynamics, on the other side of the minimum, τ_{nr}^{dim} decreases with the interaction strength.

[1] arXiv:1611.09115 [physics.chem-ph]

CPP 26.3 Tue 14:30 ZEU 260

Effective Treatment of Quantum Dissipation at a Conical Intersection — ●HONG-GUANG DUAN^{1,2,3}, R. J. DWAYNE MILLER^{1,3,4}, and MICHAEL THORWART^{2,3} — ¹Max-Planck Institute, Hamburg, Germany — ²University of Hamburg, Hamburg, Germany — ³Center for Ultrafast Imaging, Hamburg, Germany — ⁴University of Toronto, Toronto, Canada

A conical intersection (CI) is a degenerate point of two potential energy surfaces (PESs) in the configuration space of a polyatomic molecule. It enables an ultrafast radiationless transition of an electronic wave packet between two PESs. Yet, revealing the details of the nonadiabatic quantum dynamics in the vicinity of a CI is still challenging because the CI usually involves an enormous number of the electronic and nuclear degrees of freedom. Advanced quantum chemistry provide an atomic view of the quantum dynamics but limited by the number of atoms. Here, we formulated a further reduced model to describe the nonadiabatic quantum dynamics around the CI. By transforming the tuning and coupling modes into the bath, we have significantly reduced the computational complexity, which allows for an efficient numerical treatment of a CI in the presence of strong vibrational relaxation. We have calculated the two-dimensional pump-probe spectra and identify the signature of the CI. Moreover, we have studied the impact of the vibrational coherence on the quantum efficiency of the transfer via a CI. We observe that more coherent wave packets show a higher quantum yield than the less coherent one. Our observations reproduce findings from a coherent control experiment.

CPP 26.4 Tue 14:45 ZEU 260

Phase modulated femtosecond spectroscopy for weakly interacting molecular aggregates — ●ZENGZHAO LI and ALEXANDER EISFELD — Max-Planck-Institute for the Physics of Complex Systems,

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Recent phase-modulated wave packet interferometry experiments on gases of alkaline atoms showed signals from states where multiple atoms are excited simultaneously [Bruder et al., Phys. Rev. A 92, 052412 (2015)]. To shed light on experimental observations, we not only numerically solve the Schrodinger equation for a typical minimal model of two interacting particles but also develop analytical formulas based on perturbation theory. Our results indicates that phase modulated femtosecond spectroscopy can probe weak dipole-dipole interactions of molecular aggregates.

CPP 26.5 Tue 15:00 ZEU 260

Stained microparticles for lifetime encoding — ●DANIEL KAGE¹, LINN FISCHER¹, KATRIN HOFFMANN¹, MARC WITTKAMP², JENS AMESKAMP², WOLFGANG GÖHDE², THOMAS THIELE³, UWE SCHEDLER³, and UTE RESCH-GENGER¹ — ¹Bundesanstalt für Materialforschung und -prüfung (BAM), Richard-Willstätter-Str. 11, 12489 Berlin — ²Quantum Analysis GmbH, Mendelstr. 17, 48149 Münster — ³PolyAn GmbH, Rudolf-Baschant-Str. 2, 13086 Berlin

Multiparametric analyses involving optical techniques like flow cytometry are at the core of studying complex systems in biological research and diagnostic applications [1]. However, for fluorescence-based techniques, the number of reporters distinguishable in spectral multiplexing is limited by spectral overlap and requires a multitude of excitation light sources and detection systems [2]. Intensity encoding often used for bead assays suffers from problems regarding dye concentration control and excitation light intensity fluctuations [2]. An alternative is luminescence lifetime encoding [3], particularly to minimize instrument costs. Here, we report on dye-stained polymer microparticles for lifetime encoding in flow cytometry with different organic dyes. This ranges from studies of the impact of parameters like dye loading concentration and particle diameter on fluorescence decay behavior to the demonstration of lifetime code reading and simultaneous ligand fluorescence signal detection with single-wavelength excitation in a flow.

[1] V. V. Tuchin, Advanced optical flow cytometry: methods and disease diagnoses. Wiley-VCH, 2011. [2] K. Hoffmann et al., ACS Nano, 2013, 7(8), 6674. [3] R. Cao et al., Opt Express, 2013, 21(12), 14816.

CPP 26.6 Tue 15:15 ZEU 260

Following a Photocatalytic Redox Cycle using Fluorescence Correlation Spectroscopy — ●JOSEF HAIMERL¹, JAN VOGELSANG¹, INDRAJIT GHOSH², BURKHARD KÖNIG², and JOHN LUPTON¹ — ¹Institut für angewandte und experimentelle Physik, Universität Regensburg — ²Institut für organische Chemie, Universität Regensburg

Rhodamine dye-based photo-induced redox catalytic processes hold great importance as efficient and, moreover, cheap alternatives to already existing transition metal-based photocatalytic processes. Recent examples show that consecutive photo-induced electron transfer processes (conPET) overcome the energetic limit of using the energy of only a single photon in photoredox catalysis. However, often, it is not clear which excited states of the photocatalysts are involved in such processes and how solvents (e.g. solvent polarity), reductants or oxidants impact the dynamics of these systems. We therefore present the use of fluorescence correlation spectroscopy (FCS) and time-correlated single photon counting (TCSPC) in order to understand the details of such photoredox catalytic processes, as well as to characterise the steps that are crucial in those photocatalytic cycles with the corresponding efficiencies.

CPP 26.7 Tue 15:30 ZEU 260

Light-Harvesting Processes Conference Revealing The Structural Properties Of Chromophores With The Help Of MD-Simulations — ●AXEL BOURDICK and STEPHAN GEKLE — Biofluid Simulation and Modeling, University of Bayreuth, Universitätsstr. 30, 95440 Bayreuth, GERMANY

With the advent of organic photovoltaics promising materials for organic solar cells have become a strong focus of research. One particular interesting manufacture strategy is the push-pull chromophore design, whereby an electron rich donor unit and an electron deficient acceptor are placed alternating on the same molecule [1,2]. With the

help of Molecular Dynamics Simulations we investigate the Systems p-DTS(FBTTh2)2 (T1) and p-SIDT(FBTTh2)2 (H1) [3]. We are interested in the structure and conformation of aggregated states in different solvents, and physical properties, like the free energy of the system under various conditions. By comparison with experimental results we will contribute to a better understanding of the macroscopic properties of the materials in question, which can help optimizing molecular chromophore design in future applications.

[1]*D. Gendron, M. Leclerc, (2011) *Energy Environ. Sci.*, 4, 1225-1237

[2]*C. Duan, F. Huang, Y. Cao, (2012) *J. Mater. Chem.*, 22, 10416-10434

[3]*J. A. Love et al., (2014), *J. Am. Chem. Soc.*, 136(9), 3597-3606

CPP 26.8 Tue 15:45 ZEU 260

Absolute optical sensor based on biluminescence — •CATERIN SALAS REDONDO and SEBASTIAN REINEKE — Dresden Integrated Center for Applied Physics and Photonic Materials, Dresden, Germany

According to the selection rules in quantum mechanics, only transitions from the singlet excited state are allowed, deactivating radia-

tively while generating fluorescence emission, whereas transitions from the triplet excited state are theoretically not allowed. Nevertheless, there is a small probability of these forbidden transitions to occur at a low rate, resulting in a slow radiative deactivation known as phosphorescence emission. In this context, the property of an organic molecule able to emit photons from both their singlet and triplet excited states is called biluminescence. Although this dual state emission, particularly at room temperature, is difficult to achieve by purely organic molecules, it becomes possible if competitive non-radiative decay is suppressed effectively, allowing emission from the triplet states (i.e. phosphorescence) in addition to the conventional fluorescence. Here, we have identified biluminescence in simple host:guest systems in which a biluminophore is embedded in an optimum rigid matrix, for example, a combination of PMMA [poly(methyl methacrylate)] as host and NPB [N,N'-di(naphtha-1-yl)-N,N'-diphenyl-benzidine] as biluminophore. In this presentation, we will report on the oxygen sensing characteristics of a biluminescent system compared to a benchmarked single state optical sensor. Furthermore, we will address our efforts towards the potential integration of novel optical biluminescent sensing into organic electronics.