CPP 53: Hybrid, Organic and Perovskite Optoelectronics and Photovoltaics VII

Time: Friday 9:30–11:15 Location: ZEU/LICH

Invited Talk CPP 53.1 Fri 9:30 ZEU/LICH

Limits and Prospects of Organic Solar Cells — •Dieter Neher — Institute of Physics and Astronomy, University of Potsdam, Karl-Liebknecht-Str 24-25, 14476 Potsdam

Since the advent of low bandgap non-fullerene acceptors (NFAs), the performance of organic solar cells (OSCs) has improved significantly, with record efficiencies that now safely exceed 20 %. A critical parameter is the offset between the relevant frontier orbitals at the DA heterojunction, which in most NFA-based blends is the difference in HOMO energies. Here we combine a wide range of methods, from femtosecond transient absorption to steady state photoluminescence and electroluminescence, spectroscopy to study the mechanisms and efficiency of free charge generation and recombination. For a wide series of NFA-based OSCs, we find that the singlet exciton decay is the main competing pathway for free charge generation while reformation of singlet excitons from reformed CT states dominates the radiative recombination in EL. To explain our data as function of the HOMO offset, we set up a 5-state model which includes singlet and triplet excitons. Our results show that state-of-the-art OSCs already exhibit an optimum HOMO-HOMO-offset. Future work should, therefore, focus on optimizing photon harvesting and reducing bimolecular recombination in order to push the device efficiency well beyond 20 %.

CPP 53.2 Fri 10:00 ZEU/LICH

Temperature Degradation of Slot-Die Printed Organic Solar Cells — •Christoph G. Lindenmeir^1, Simon A. Wegener^1, Christopher R. Everett^1, Julian E. Heger^1, Sigrid Bernstorff^2, and Peter Müller-Buschbaum^1 — ^{1}TUM School of Natural Sciences, Chair for Functional Materials, Garching, Germany — ^{2}Elettra-Sincrotrone Trieste S.C.p.A., Basovizza, Trieste, Italy

Organic solar cells (OSCs) have emerged as promising candidates for next-generation photovoltaics due to their rapid efficiency improvements, low toxicity, and compatibility with low-cost, solution-based fabrication methods such as slot-die printing. Their high optical absorption, combined with their lightweight and flexible design, makes them particularly attractive for space applications. Offering a superior power-to-weight ratio compared to conventionally used III-V multijunction cells, OSCs can significantly reduce manufacturing and launch costs. While short-term demonstrations have confirmed their functionality in space, their long-term stability under extreme temperatures, high vacuum, and ionizing radiation still needs to be investigated. Therefore, this study compares the influence of constant-temperature conditions on active-layer morphology and electrical performance with that of temperature cycling. To investigate the degradation process, we employed operando grazing-incidence small-angle X-ray scattering (GISAXS), a nondestructive technique for probing thin-film morphology. Simultaneously, we monitored the electrical performance of the devices under vacuum to simulate space conditions.

CPP 53.3 Fri 10:15 ZEU/LICH

Electronic coarse-graining for accurate and scalable modeling of organic semiconductors — •Andriy Zhugayevych^1, Denis Andrienko^1, and Sergei Tretiak^2 — ^{1}Max Planck Institute for Polymer Research — ^{2}Los Alamos National Laboratory, USA

Modeling the electronic properties of organic semiconductors remains challenging due to their complex multiscale structure, which limits the applicability of methods developed for small-molecule solids and inorganic crystals. Fragmentation techniques designed for biomolecular systems are unsuitable for π-conjugated materials because of wavefunction delocalization. We present an electronic coarse-graining methodology that enables accurate and scalable electronic-structure calculations for organic semiconductors composed of monomers with closed-shell π-systems, allowing complete fragmentation along σ-bonds. The approach is demonstrated for crystalline polymers, highlighting how side-group modifications influence the electronic properties of thiophene-based systems such as polythiophene, P3HT, and PEDOT [1]. We further analyze errors introduced by oversimplified electronic models using examples of large flexible molecules, including non-fullerene acceptors for solar cells [2] and OLED materials. The approach can be applied - albeit in less automated way - to broader class of semiconductors, including metal-organic polymers [3], frameworks, and some inorganic semiconductors [4].

[1] Preprint at https://cmsos.github.io/escp; [2] J. Chem. Phys. 159, 024107 (2023); [3] Chem. Sci. 13, 8161 (2022); [4] J. Phys. Chem. Lett. 12, 4674 (2021).

CPP 53.4 Fri 10:30 ZEU/LICH

Facile Correction Method for DFT Band Structures Towards Hybrid Density Functional Level for 2D-Covalent Organic Frameworks — •Laura Fuchs, Maximilian F. X. Dorfner, Matthias Knechtges, Johann Olbrich, and Frank Ortmann — Department of Chemistry, TUM School of Natural Sciences, and Atomistic Modeling Center, Munich Data Science Institute, Technical University of Munich, Germany

Density functional theory (DFT) is the method of choice for predicting the majority of structural, and electronic properties of organic and inorganic materials. Generalized gradient approximations, such as the semi-local PBE functional, are widely used in the community but have one major drawback: the band gap problem. The fundamental band gap is typically underestimated by up to a factor of two by these approaches, which is critical since this electronic property is of utmost importance for assessing the applicability of the investigated material for (opto-)electronic purposes.

Here, we establish a robust linear correlation between the semi-local PBE band gaps and the more accurate, but more expensive, hybrid HSE06 band gaps by analyzing organic molecules as well as extended crystals such as covalent organic frameworks (COFs). By eliminating the need for costly hybrid functional calculations, our approach achieves up to a 54-fold speedup compared to conventional HSE06 computations. We compare various implementation strategies and evaluate each method's advantages and limitations, with particular focus on their implications for predicting electronic transport properties.

CPP 53.5 Fri 10:45 ZEU/LICH

The influence of vibrations on charge and energy transfer dynamics in a singlet fission donor-acceptor complex — •Karin S. Thalmann^1, Johan E. Runeson^1, Pedro B. Coto^2, and Michael Thoss^1 — ^{1}Institute of Physics, University of Freiburg, Germany — ^{2}Spanish National Research Council (CSIC), Madrid, Spain

Singlet fission is a photophysical process in molecular materials transforming a singlet excited electronic state to two triplet states [1], effectively doubling the number of charge carriers. This makes molecules exhibiting singlet fission possible candidates to increase the efficiency of solar cells beyond the Shockley-Queisser limit. To investigate this possibility, we analyse the charge and energy transfer dynamics in a donor-acceptor complex consisting of a bis(diazadiborine)-based chromophore [2] and tetracyanoquinodimethane. Using a combined approach of *ab initio* multireference perturbation theory calculations and quantum dynamical simulations based on a harmonic vibronic coupling Hamiltonian [3], we reveal competing charge and energy transfer mechanisms, such as singlet fission-based transfer and energy decay channels. Further, we expand our model Hamiltonian to include all vibrational degrees of freedom and the anharmonicity in the vibrational modes of the chromophore. The use of mixed quantum-classical approaches allows us to analyse the influence of these vibrational modes on the dynamics as well as the charge and energy transfer mechanisms.

[1] M. B. Smith *et al.*, *Chem. Rev.* **110**, 6891-6936 (2010).
[2] T. Zeng, *J. Phys. Chem. Lett.* **7**, 4405-4412 (2016).
[3] S. R. Reddy *et al.*, *J. Chem. Phys.* **151**, 044307 (2019).

CPP 53.6 Fri 11:00 ZEU/LICH

Tracking the degradation of non-fullerene organic solar cells in fluctuating environments — •Lixing Li, Thomas Baier, Xiaojing Ci, Zhaonan Jin, and Peter Müller-Buschbaum — TUM School of Natural Sciences, Chair for Functional Materials, 85748 Garching, Germany

The long-term operational stability of non-fullerene organic solar cells (OSCs) remains a critical challenge that limits practical applications. While substantial efforts have been devoted to understanding the degradation mechanisms of OSCs under extreme conditions such as high or low temperatures, far less attention has been paid to their potential recovery behaviour. In real-world environments, organic solar cells are not exposed to constant stress; instead, environmental factors such as temperature, humidity, and illumination fluctuate continuously over time. These dynamic variations may induce partial or even reversible changes in device performance and morphology. It is essential to explore not only how OSCs degrade under stress but also how they recover once the stress is relieved or altered. In this study, we investigate the recovery processes of BTP-4F non-fullerene organic solar cells subjected to multiple environmental parameters. Advanced characterisation techniques, including grazing-incidence X-ray scattering (GIXS) and atomic force microscopy (AFM), are employed to track both in-situ and ex-situ structural evolution within the active layer. These analyses provide new insights into the mechanisms governing the recovery and structural reorganisation of OSCs under dynamic environmental conditions.