CPP 25: Hybrid, Organic and Perovskite Optoelectronics and Photovoltaics II

Time: Tuesday 11:30–12:45 Location: ZEU/0260

CPP 25.1 Tue 11:30 ZEU/0260

Transiently Delocalised Hybrid Quantum States are the Gateways for Efficient Exciton Dissociation at Organic Donor-Acceptor Interfaces — •Filip Ivanovic[1], Samuele Giannini[2], Wei-Tao Peng[3], and Jochen Blumberger[1] — [1]Department of Physics and Astronomy, University College London, London, United Kingdom — [2]Department of Chemistry and Industrial Chemistry, University of Pisa, Pisa, Italy — [3]Department of Chemistry, Tunghai University, Taichung City, Taiwan

Organic solar cells (OSCs) have long been the subject of intensive research. Following the absorption of sunlight, localised excitations (excitons) are able to dissociate into separated charges at the interface between donor and acceptor components. Whilst this separation has been leveraged to achieve cell efficiencies above 20%, the exact mechanism by which this occurs remains without consensus.

Theoretical research has been centred on simulating such phenomena on experimentally relevant time scales, whilst sufficiently describing the underlying electronic structure. Here, we introduce our in-house non-adiabatic dynamics package, termed X-SH.

We elucidate the mechanism of ultrafast charge separation in an atomistic OSC junction, where separation is mediated by non-local hybridised states comprising both excitons and separated charges, and in-fact accelerated by non-interfacial exciton dissociation. We then demonstrate that tuning the hybridised states' accessibilities through the density of states can be translated into design rules informing the synthesis of yet more efficient OSCs.

CPP 25.2 Tue 11:45 ZEU/0260

Temperature-Dependent Spin-Sensitive Spectroscopy of Triplet-Exciton Dynamics in TADF OLEDs — •Ahmed Mohamed, Andreas Kohrmann, Vladimir Dyakonov, and Andreas Sperlich — Experimental Physics 6, University of Würzburg, 97074 Würzburg, Germany

Thermally Activated Delayed Fluorescence (TADF) enables efficient harvesting of triplet excitons through reverse intersystem crossing (rISC) in OLEDs. Previous studies have shown that TADF device operation is dominated by triplet excitons, whose densities exceed singlet and charge carrier densities by several orders of magnitude. A major limitation to TADF efficiency is the non-radiative loss of triplet excitons. Here, we analyze the transient photoluminescence (trPL) and transient electroluminescence (trEL) decays of the blue TADF emitter ν-DABNA hosted in TDBA-Si or mCP. Temperature-dependent trPL/trEL kinetics were modeled using a rate-equation framework. Key triplet-exciton parameters, including the lifetime, diffusion coefficient, diffusion length, and activation energies were extracted by considering both 1^{st} and 2^{nd} order triplet-exciton decay pathways. The spin properties of the triplet exciton were additionally identified using photoluminescence-detected magnetic resonance (PLDMR) measurements on the organic layer stack. Together, kinetic and spin analysis of triplet excitons provide a comprehensive picture of non-radiative quenching to guide future improvements of TADF OLEDs

CPP 25.3 Tue 12:00 ZEU/0260

Modeling the ionization potential and electron affinity in organic semiconductors — •Alexey Gudovannyy and Frank Ortmann — Department of Chemistry, TUM School of Natural Sciences, and Atomistic Modeling Center, Munich Data Science Institute, Technical University of Munich, Germany

Ionization potential (IP) and electron affinity (EA) are essential quantities for characterizing and tuning transport properties of organic semiconducting materials. Computational prediction of these properties under realistic conditions of crystalline or polycrystalline forms remains challenging, as it requires a proper description of all interactions within the bulk material environment. In this work, we present an accurate protocol for computing molecular materials' IP and EA values, using computed gas-phase molecular values corrected via an improved multilevel scheme for describing the materials' environment, where an electronic structure is treated explicitly at the microscopic level, while at larger scales, a continuum model is applied. Special significance will be dedicated to the requirement of accurate reference gas-phase calculations, which we carried out within the GW framework and to demonstrating limitations of prevalent Density Functional Theory (DFT) methods, while preserving feasible computational time demands. The main bottleneck of this approach is the need for an experimental or predicted crystal structure. We will also show how to incorporate reliable structure prediction into it, which opens up possibilities for purely in silico materials design.

CPP 25.4 Tue 12:15 ZEU/0260

Morphological Insights into Spray-Coated Organic Semiconductors — •Shuxian Xiong[1,2], Marie Betker[2,3], Simon Schraad[1,2], Yufeng Zhai[2], Benedikt Sochor[2], Sarathlal Koyiloth Vayalil[2,4], Peter Müller-Buschbaum[1], and Stephan V. Roth[2,3] — [1]TUM School of Natural Sciences, Chair for Functional Materials, Garching, Germany — [2]Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany — [3]Department of Fibre and Polymer Technology, KTH, Stockholm, Sweden — [4]Applied Sciences Cluster, University of Petroleum and Energy Studies UPES, Dehradun, Uttarakhand, 248007, India

Ultrasonic spray coating offers improved scalability for large-scale applications, reduced material waste, and enhanced compatibility with various substrates, making it a more cost-effective solution for the mass production of photovoltaic devices. However, how to simply and efficiently build ordered structural networks is still a critical issue. We investigate the morphological transition of organic semiconductors and their effect on device performance using grazing incidence wide-angle X-ray scattering in traditional fullerene systems. By systematically optimizing spray flow rate and substrate temperature, we effectively tailored the film morphology, enhancing molecular crystallization of PTB7-Th and PCBM, resulting in an improvement in device performance of 9% in air, which is one of the highest device efficiencies currently achieved for spray-coated fullerene acceptor-based organic solar cells, and is equivalent to the device efficiencies of spin-coated fullerene acceptor-based organic solar cells.

CPP 25.5 Tue 12:30 ZEU/0260

Enhancing device performance parameters of organic photodiodes by understanding the thickness dependence of the reverse dark current — •Fred Kretschmer[1], Tianyi Zhang[1], Oskar J. Sandberg[2], Mathias Nyman[2], Karl Leo[1], and Johannes Benduhn[1] — [1]Institute of Applied Physics, TU Dresden, Dresden, Germany — [2]Faculty of Science and Engineering, Åbo Akademi University, Turku, Finland

In recent years, organic photodetectors (OPDs) have attracted interest due to their advantageous properties and rapidly advancing performance, making them promising not only as complements to inorganic photodetectors but also for emerging applications in bioimaging, health monitoring, and high-speed optoelectronics. A key figure of merit for comparing PDs is the specific detectivity (D^*), which quantifies the detector's ability to sense weak optical signals. Achieving high D^* relies on maximizing charge carrier extraction while minimizing internal noise. Under reverse bias, OPD performance is particularly influenced by shot noise, which is proportional to the dark current (J_D).

In this work, the variation of J_D with active-layer thickness across multiple energy-gap-tuned bulk-heterojunction systems was examined. Surprisingly, the dark current exhibits a thickness-dependent minimum, contrary to the general assumption that J_D only decreases with thickness. Using sensitive, temperature-dependent current-voltage measurements together with drift-diffusion simulations, the new minimum feature was confirmed, and improvements in the specific detectivity of OPDs were achieved.