

CPP 18: Organic Electronics and Photovoltaics I (joint session CPP/HL)

Time: Tuesday 9:30–13:00

Location: GÖR 226

CPP 18.1 Tue 9:30 GÖR 226

Use of a multiple hydride donor to achieve an n-doped polymer with high solvent resistance — FARZANEH SAEEDIFARD^{1,2}, DOMINIQUE LUNGWITZ³, ZI-DI YU⁴, SEBASTIAN SCHNEIDER⁵, AHMED E. MANSOUR^{3,6}, ●ANDREAS OPITZ³, STEPHEN BARLOW^{1,2}, MICHAEL F. TONEY¹, JIAN PEI⁴, NORBERT KOCH^{3,6}, and SETH R. MARDER^{1,2} — ¹University of Colorado Boulder, US — ²Georgia Institute of Technology, Atlanta, US — ³Humboldt-Universität zu Berlin, Germany — ⁴Peking University, China — ⁵Stanford University, US — ⁶Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Germany

Insolubilization of doped semiconducting polymer layers can help to fabricate efficient multilayer solution-processed electronic and optoelectronic devices. Here, we present a promising technique to simultaneously n-dope and largely insolubilize conjugated polymer films using tetrakis[4-(1,3-dimethyl-2,3-dihydro-1*H*-benzo[*d*]imidazol-2-yl)phenoxy]methane (tetrakis-O-DMBI-H), which consists of four 2,3-dihydro-1*H*-benzoimidazole (DMBI-H) n-dopant moieties covalently linked to one another. [1] Doping a thiophene-fused benzo[*d*]furandione-based oligo(*p*-phenylenevinylene)-*co*-thiophene polymer (TBDOPV-T) with tetrakis-O-DMBI-H results in a highly n-doped film with bulk conductivity of 15 S/cm. Optical absorption spectra reveal a film retention of ~93% after immersion in *o*-dichlorobenzene for 5 min. This is caused by multiple electrostatic interactions the multiple electrostatic interactions between each dopant tetra-cation and up to four nearby anionic doped polymer segments.

[1] F. Saeedifard et al., *ACS Appl. Mater. Interfaces* **14** (2022) 33598.

CPP 18.2 Tue 9:45 GÖR 226

Numerical Simulation of Crystallization Kinetics in Binary Mixtures for Organic Photovoltaic Applications — ●MAXIME SIBER, OLIVIER RONSIN, and JENS HARTING — Helmholtz-Institute Erlangen-Nürnberg for Renewable Energies (HI-ERN), Germany

With the aim to understand the formation process of organic photoactive layers, and identify process-structure relationships in order to optimize their fabrication, a computational framework relying on the phase-field modelling approach has recently been developed. It is now sought to quantitatively compare numerical simulations of drying organic photovoltaic (OPV) films with corresponding experimental realizations.

Since the fabrication process involves the interplay of multiple physical phenomena, which separate characterization already is a challenging task from the simulative, as well as from the experimental side, this first study focusses on crystallization behaviour in isothermal, non-evaporating, binary OPV blends. In particular, this talk addresses crystalline structures that arise from different crystallization processes such as solution-triggered crystallization, diffusion-limited crystallization, or spinodal decomposition-assisted crystallization. Furthermore, crystallization kinetics quantified for several blend ratios and material parameters are also analyzed and compared. Finally, perspectives regarding validation against in-situ experiments and derivation of design rules for OPV film fabrication are also provided.

CPP 18.3 Tue 10:00 GÖR 226

Optical properties of perfluorotetracene (PFT) crystal polymorphs — ●ANA MARIA VALENCIA^{1,2} and CATERINA COCCHI^{1,2} — ¹Institut für Physik, Carl von Ossietzky Universität Oldenburg — ²Physics Dept., Humboldt-Universität zu Berlin und IRIS Adlershof

The family of perfluorinated acenes is gaining popularity among organic materials for optoelectronics. Since the electronic and optical properties of molecular solids are crucially dependent on their packing motifs, it is crucial to consider crystalline phases in the simulations to understand the structure-property relationships of these materials, especially regarding electronic correlations and excitations. Here, we investigate from first principles by means of density functional theory and many-body perturbation theory the electronic structure and the optical excitations of two crystal polymorphs of perfluorotetracene, considering for comparison tetracene and the corresponding isolated molecules. This way, we can assess the effects of fluorination as well as of the crystal periodicity. We find that the absorption spectra are only mildly influenced by the packing motif. However, our analysis gives insight into the exciton binding energies as well as the spatial distribution of the excitons. We inspect not only the first excitations but also

the higher-energy ones, thus providing an all-around understanding of the optical excitations in these materials.

CPP 18.4 Tue 10:15 GÖR 226

Controlling long-range order in dip coated piezo- and ferroelectric molecular materials — ●ANDREY BUTKEVICH and MARTIJN KEMERINK — Institute for Molecular Systems Engineering and Advanced Materials, Im Neuenheimer Feld 225, 69120 Heidelberg, Germany

Organic piezo- and ferroelectrics are of great interest due to their tunable properties. However, the extensive procedures that are often required to achieve saturation polarization limit their possible applications. A so far unexplored possible solution is to pre-align the molecules during the deposition from solution. Here, we analyzed the effect of dip coating parameters on the alignment of multiple supramolecular piezo- and ferroelectrics. Dip coated films were characterized for different material concentrations and dip coating velocities. The investigation revealed that morphologies with strong alignment along the dip coating direction are formed for both material types. For ferroelectrics, the changes in the ferroelectric switching behavior of the resulting thin films were investigated. The ferroelectric switching was investigated using the double wave method, revealing that dip coating perpendicular to the electrodes leads to an almost perfect in-plane alignment of the supramolecular polymers, which was confirmed by X-ray diffraction. The observed switching behavior was comparable to films that were aligned via conventional field-annealing. Hence, dip coating enhanced the ferroelectric switching in the investigated molecules, which we anticipate can be directly transferable to other molecular ferroelectrics.

CPP 18.5 Tue 10:30 GÖR 226

In search of novel organic ferroelectrics — ●HEIKO MAGER — IMSEAM, Heidelberg University

The switchable polarization and concomitant pyroelectric and piezoelectric properties of ferroelectric materials enable a variety of applications, ranging from memory devices over thermal and mechanical sensors to energy harvesters. Although current applications are dominated by inorganic materials, organic ferroelectrics offer a flexible, cheap and possibly non-toxic and biodegradable alternative.

Here, we present experimental research on the ferroelectric-like behavior of the novel liquid crystalline molecule C₆H₆F₅O-C₃-Amide and its molecular derivatives. To screen for a possible para- to ferroelectric phase transitions, dielectric spectroscopy was employed. While the for ferroelectric materials characteristic Curie-Weiß behavior was not directly observed, features in the dielectric suggest a possible phase transition coinciding with a peak in differential scanning calorimetry traces.

Polarization hysteresis loops were obtained via double wave measurements and characteristic capacitance-voltage "butterfly" loops measured. Comparisons between the different molecular derivatives allow insights into the molecular groups relevant for the switching process.

CPP 18.6 Tue 10:45 GÖR 226

Implementation and simulation of drift-diffusion models for organic mixed conductor devices — ●ANDRES DAVID PEÑA UNIGARRO^{1,2} and FLORIAN STEFFEN GUNTHER^{2,3} — ¹Institute of physics, TU Chemnitz, Chemnitz, Germany — ²IFSC, University of São Paulo, Brazil — ³UNESP, Rio Claro, Brazil

Organic electrochemical transistors (OECTs) have emerged as potential transducers in applications that require the conversion of ion fluxes to electronic current. For the understanding of the fundamental mechanism in OECTs and OECT-based applications, as well as for their rational optimization, however, it is essential to have theoretical models that agree with experimentally measured device responds. Most of the existing OECT models consider that the ion flux from the electrolyte into the organic, semiconducting layer takes place only due to an electrical field. These models are efficient to describe the steady state operations of OECTs, but are rather limited when compared to transient behaviors. Recently, more refined models which take a diffusion term into account, have been developed. The simplifications

needed to use an analytical solution of the governing equation, however, reduces its generality, limiting the results to specific cases. In our work, we use numerical methods to solve the drift-diffusion equation in one dimension to overcome these limitations of the analytical solution. This allows us to go beyond the standard boundary conditions and to analyze the impact of other alterations.

CPP 18.7 Tue 11:00 GÖR 226

Noise in Organics Semiconductors and Devices — ●PRIYA VIJI, DOROTHEA SCHEUNEMANN, CONSTANTIN TORMANN, and MARTIJN KEMERINK — Institute for Molecular Systems Engineering and Advanced Materials, Heidelberg University, Germany

Noise, typically considered an unwanted signal in measurements, can also contain information about the charge kinetics in organic semiconductors. Since the corresponding devices often show high resistances and capacitances, the measurement of –albeit very small– current fluctuations via voltage noise is most suitable. To quantify this noise and to eliminate noise from other devices in the measurement line, a cross-correlation technique is implemented and applied to P3HT doped with varying concentrations of F4TCNQ. The results compare quantitatively with kinetic Monte Carlo simulations in which noise calculations were implemented. To understand the effect of the electric field on the charge carriers, disentangling noise in directions parallel and perpendicular to the electric field is essential. The direction perpendicular to the electric field does show a gradual increase at high electric fields, which matches the concept of an Effective Temperature, as described by Marianer and Shklovskii, in which the effects of electric field and lattice temperature are combined. In the direction of the electric field, an additional contribution due to shot noise is observed. However, its magnitude deviates from the theoretical prediction of shot noise, which can be quantified as a non-unity Fano-factor.

15 min. break

CPP 18.8 Tue 11:30 GÖR 226

Uniaxially Aligned Merocyanine Films by Graphene Nanoribbon Templated Growth — ●PHILIPP WEITKAMP, LUKAS BÖHNER, NORA GILDEMEISTER, DIRK HERTEL, and KLAUS MEERHOLZ — Physikalische Chemie, Universität zu Köln, Deutschland

We herein report the unique and novel approach of achieving a polycrystalline thin film consisting of uniaxially aligned domains by using 7-armchair graphene nanoribbon (7-aGNR) monolayers as van-der-Waals template. For this purpose, a merocyanine dye was evaporated on 7-aGNRs, transferred on quartz glass substrates. The alignment of the formed molecular aggregate along the GNR alignment direction was proven by polarisation dependent absorbance spectroscopy. The J- and H-transition, formed by the dye aggregate, were correlated with distinct axes of the crystal structure. By combining this correlation with polarisation dependent absorbance measurements and X-ray diffraction experiments we elucidated the three-dimensional structure of the formed aggregate thin film. The growth mode of these films was investigated as a function of the applied layer thickness. Atomic force microscopy-based morphology analysis and X-ray diffraction experiments were used to reveal the anisotropic on-surface crystallisation along the 7-aGNR long axes direction. Furthermore, we found that the delocalisation length of the aggregate increases with increasing in-plane order. Finally, we demonstrated that the in-plane alignment leads to an anisotropic charge carrier transport by implementing the templated merocyanine thin film as active layer in a top-gated organic field effect transistor.

CPP 18.9 Tue 11:45 GÖR 226

Investigation on organic light-emitting diodes, fabricated by a self-developed and highly automated physical vapor deposition system — ●FABIAN BINDER, MONA LÖTHER, PASCAL SCHADY, VLADIMIR DYAKONOV, and ANDREAS SPERLICH — Experimental Physics VI, Julius Maximilian University of Würzburg, 97074 Würzburg

Organic light emitting diodes (OLEDs) are most commonly produced in ultra-high vacuum by physical vapor deposition (PVD) of several organic and metallic layers on a carrier substrate. We developed an automated PVD system, mainly consisting of two vacuum chambers for organic and metal deposition, respectively. Stepper motors and sensor solutions ensure a smooth and precise positioning of the carrier substrate above the evaporation crucibles. The substrate is then rotated with a defined speed to achieve an even deposition of the ma-

terial. In order to vapor-deposit material, a certain material-specific temperature range is required which is realized by a software-based temperature controller which manages the evaporation rate according to the user's specifications. A user interface facilitates planning of the desired OLED layer stack and provides information about the production progress. The capabilities of our new PVD system were tested by producing OLEDs based on multiple-resonance TADF emitters of the DABNA series. We performed electro-optical LJV-characterization and determined the devices' quantum efficiencies. The results of this investigation will enable us to optimize the production process with regard to evaporation rates and layer thicknesses.

CPP 18.10 Tue 12:00 GÖR 226

improvement of electrical transport in organic semiconductor thin films by charge transfer doping — ●HONGWON KIM¹, ANDREAS OPITZ², FLORIAN FENZL¹, and WOLFGANG BRÜTTING¹ — ¹Experimentalphysik IV, Institut für Physik, Universität Augsburg — ²Institut für Physik, Supramolekulare Systeme, HU zu Berlin

Charge carrier transport in organic semiconductor is based on the hopping mechanism. We have used in-situ charge transfer doping during film-growth to improve the electrical conductivity by simultaneously increasing the carrier density and reducing their thermal activation energy. By doping planar organic semiconductors, such as DBTTF, a-6T, and DIP, with strong acceptors (F6-TCNNQ & HATCN), hybridization of pi-orbitals occurs while generating new interphases (charge transfer complex, CTC). On the other hand, DBP, a non-planar molecule, forms an amorphous thin film without any CTC generation. To measure the characteristics of CTC, we used near-infrared transmission spectroscopy, which can detect the absorption peaks of CTCs and the energy gap. In addition, the activation energy is obtained from temperature-dependent conductivity measurements. We observe a correlation between the magnitude of the electrical conductivity and its activation energy, which can be ascribed to the binding energy of CTCs. Depending on the specific system, conductivity maxima are observed between 10 and 30% molar doping ratios.

CPP 18.11 Tue 12:15 GÖR 226

Electrical transport dynamics of conducting polymers in nanoconfinement — SUKANYA DAS and ●K.S. NARAYAN — Chemistry and Physics of Materials Unit, Jawaharlal Nehru Centre for Advanced Scientific Research, Bengaluru- 560064, India

Inherently disordered conducting polymers consist of different characteristic lengths, defect distribution over the polymer chains and their microstructural modifications can alter the transport properties significantly. The electrical transport of conducting polymers such as poly(3,4-ethylene dioxythiophene): poly(styrenesulfonate) PEDOT:PSS has shown conductivity enhancement when confined in an array of nanochannels in the form of alumina nanopillars. The transverse conductivity along the nanopillar axis, which is an otherwise suppressed quantity, increases nonlinearly by approximately three orders in magnitude as the diameter of the scaffold-channel is decreased to 20 nm, when measured at single nanochannel level as well as at bulk-macroscopic level. This suggests the major role of dimensions and geometry in eliciting efficient electrical transport. Similarly, more than 3 orders of conductivity increase have been shown by in-situ polymerized poly(3,4-ethylenedioxythiophene):tosylate within the nanochannels which has similar electronic backbone PEDOT but a different dopant environment. The microscopic insight into PEDOT:Tos has shown higher degree of crystallinity and ordering as compared to PEDOT:PSS nanochannels. We utilize this property of PEDOT:PSS to fabricate hybrid organic-inorganic perovskite photodiode with efficient hole extraction and faster response times in the nanoconfined devices.

CPP 18.12 Tue 12:30 GÖR 226

In-situ spectro-electrochemistry of N-type conjugated organic systems for opto-electronic and electro-optic applications — ●MEENAL KATARIA and SABINE LUDWIGS — IPOC-Functional Polymers, Institute of Polymer Chemistry, University of Stuttgart, Pfaffenwaldring 55, 70569 Stuttgart, Germany

This talk will highlight our ongoing research on in-situ spectroelectrochemistry. We are developing functional organic conjugated materials and devices for opto-electronic and electro-optic applications. Here, one of our aims is to utilize an in-situ spectroelectrochemical approach to calculate the energy levels of blends of donor polymers (e.g. PM6) and non-fullerene acceptors (e.g. Y6) utilized for high performance organic solar cells. For relevant device information it is important to have precise values of energy levels of the same blend systems as used

in devices. Thus, our absorption assisted electrochemistry technique proved be advantageous approach (1). In another research work discussing the effect of isomerism on conductivity of n-type polymers (e.g. P(NDI2OD-T2)), this above approach has been used to identify the redox states during electrochemical doping which helped to identify the charge transfer transport mechanism (2). In the last example, this technique has been utilized to calculate the frontier energy levels of push-pull D- π -A systems and to study the effect of acceptor strength on their energy levels (3).

Ref. (1) J. Mater. Chem. C 2022, 10, 11565. (2) Chem. Mater. 2019, 31, 3542. (3) Phys.Chem.Chem.Phys. 2020, 22, 2283.

CPP 18.13 Tue 12:45 GÖR 226

Delocalization Enhances Conductivity at High Doping Concentrations — •DENNIS DEREWJANKO¹, DOROTHEA SCHEUNEMANN¹, EMMY JÄRSVALL², ANNA I. HOFMANN², CHRIS-

TIAN MÜLLER², and MARTIJN KEMERINK¹ — ¹IMSEAM, Heidelberg University, Im Neuenheimer Feld 225, 69120 Heidelberg, Germany — ²Department of Chemistry and Chemical Engineering, Chalmers University of Technology, 41296 Gothenburg, Sweden

Many p-type organic semiconductors are experimentally found to follow a universal power-law trend between conductivity and charge carrier concentration at practically relevant high doping levels. This behavior cannot consistently be explained by conventional charge transport models. Here, we develop a physically transparent model based on the combination of a tight binding model and a variable range hopping model to show that the observed power-law trend can be explained by consideration of an energy dependent localization length. The underlying cause is an energetic lifting of the charge carriers to partly delocalized states due to the rising Fermi energy level at high charge carrier concentration. At low charge carrier concentrations, the well-known Mott-Martens model is recovered.