

## CPP 55: Focus: Static and Dynamic Disorder Phenomena on the Transport in Organic Semiconductors I

Organisers: Jens Pflaum (Universität Würzburg), Thomas Kirchartz (FZ Jülich), Dieter Neher (Universität Potsdam)

Transport of charge carriers as well as of excitons defines a crucial process in the successful implementation of organic semiconductors in modern thin-film applications. In particular, structural and energetic disorder influences the localization of charge carriers and excitons and, thus, exerts a tremendous influence on the nature of transport in small molecule as well as polymer semiconductors. However, as demonstrated by recent theoretical work, the correlation between disorder and charge transport becomes much more complex taking into account dynamic effects and their related time-scales. In certain cases, this might even lead to delocalization of previously localized charge carriers, thereby being beneficial for the overall macroscopic transport. Therefore, this focus session aims for bringing together experimentalists as well as theoreticians working in the field of charge and exciton transport in organic materials. Only by complementary contributions from different areas in organic solid states, including rational design of materials, development of advanced methods and state-of-the-art calculations, a comprehensive understanding of the relationship between disorder and transport will be feasible and, thus, a successful application of advanced optimization strategies to real devices. As such, this focus session is intended to provide the suited platform for an intense informational exchange on this topic and, thereby, to initiate possible future collaborations between the relevant groups.

Time: Thursday 10:15–13:00

Location: ZEU 222

### Invited Talk

CPP 55.1 Thu 10:15 ZEU 222

**Understanding charge transport in crystalline organic semiconductors** — ●SIMONE FRATINI<sup>1</sup>, SERGIO CIUCHI<sup>2</sup>, ALESSANDRO TROISI<sup>3</sup>, and DIDIER MAYOU<sup>1</sup> — <sup>1</sup>Institut Néel - CNRS and Université Grenoble Alpes, Grenoble, France — <sup>2</sup>Department of Physical and Chemical Sciences, University of L'Aquila & CNR-ISC, L'Aquila, Italy — <sup>3</sup>Department of Chemistry, University of Warwick, UK

Charge transport in crystalline organic semiconductors is intrinsically limited by the presence of large thermal molecular motions, which are a direct consequence of the weak van der Waals intermolecular interactions. These lead to an original regime of transport called transient localization, involving the quantum localization of the carriers at timescales shorter than the typical molecular motions. After a brief review of experimental observations that pose a challenge to textbook theories, I will present model calculations supporting the transient localization scenario. I will show that the theory consistently explains several common experimental features of organic semiconductors, such as the low mobilities falling below the Mott-Ioffe-Regel limit, the typical power-law temperature dependence observed in clean crystalline compounds as well as the observed non-Drude like behavior of the optical conductivity. I will then discuss efficient strategies to improve the mobility in this important class of materials and conclude with a systematic screening performed on the whole family of organic semiconductors, showing how the electronic structure determines their sensitivity to molecular disorder. Mobilities above 100 cm<sup>2</sup>/Vs are predicted to occur in appropriately tailored compounds.

### Invited Talk

CPP 55.2 Thu 10:45 ZEU 222

**Mapping of trap densities and contact resistance in organic devices and their relation to structural disorder** — ●BERT NICKEL<sup>1</sup>, CLEMENS LIEWALD<sup>1</sup>, SIMONE STROHMAYER<sup>1</sup>, ERIC GLOWACKI<sup>2</sup>, and ANDREY TURCHANIN<sup>3</sup> — <sup>1</sup>Ludwig-Maximilians-Universität, Department of physics & CeNS, D-80539 Munich, Germany — <sup>2</sup>Linköpings Universitet, Laboratory of Organic Electronics, SE-601 74 Norrköping, Sweden — <sup>3</sup>Friedrich-Schiller-Universität Jena, Institute of Physical Chemistry, 07743 Jena, Germany

We develop experimental tools to explore the influence of static and dynamical disorder phenomena on transport in organic devices. Examples studied range from simple aromatics such as pentacene and DNNT to H-bonded pigments such as epindolidione. We aim on correlating the electronic transport properties [1] with local structural properties obtained from nano beam x-ray diffraction experiments and near field microscopy [2]. Photocurrent microscopy allows us to determine local transport properties such as trap densities and charge carrier dynamics in fully operational thin film transistors, as well as local distribution of contact resistances. Contact resistance is often related to imperfect growth on metal contacts, we will present new approaches to deposit highly ordered films by transfer techniques.

[1] C. Liewald, D. Reiser, B. Nickel, Applied Physics Letters 109,

p53301 (2016)

[2] C. Westermeier, A. Cernescu, S. Amarie, C. Liewald, F. Keilmann, B. Nickel, Nature Communications 5, Article Number 4101 (2014)

### 15 min break

CPP 55.3 Thu 11:30 ZEU 222

**Construction of Polycrystalline Morphologies: A Case Study for C60 and Pentacene** — ●SEBASTIAN SCHELLHAMMER<sup>1,2,3</sup>, FRANK ORTMANN<sup>1,2</sup>, and GIANAURELIO CUNIBERTI<sup>1,2,3</sup> — <sup>1</sup>Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden — <sup>2</sup>Dresden Center for Computational Materials Science, TU Dresden — <sup>3</sup>Center for Advancing Electronics Dresden, TU Dresden

Based on the processing conditions, the morphology of small-molecule organic semiconducting materials can be varied from highly ordered single crystals to polycrystalline and amorphous. Also grain shapes vary from spherical, platelets, or rod-like. Differences in the morphology affect the energy landscape and, thus, the macroscopic charge transport behavior of the film. We present a nucleation-equilibration approach for the construction of morphologies with gradually varying degree of structural ordering. Based on geometrical considerations, the computational requirements are low enabling the construction of systems of experimentally relevant sizes. Application is demonstrated for C60 and pentacene structures varying from almost amorphous to highly ordered phases. We will also discuss the impact of morphologies on charge transport.

CPP 55.4 Thu 11:45 ZEU 222

**Dithienylethene as Switchable Hole Trap in Organic Semiconductors** — ●TIM SCHAEFER, DIRK HERTEL, and KLAUS MEERHOLZ — Department of Chemistry, University of Cologne, Germany

Trap states can considerably hinder charge carrier transport. To better understand the influence of traps on the charge transport properties of organic semiconductors, we mix in traps on purpose. We present a unique hole trap, dithienylethene (DTE), which can be photochemically switched "off" [1] to tune the trap concentration in one and the same device. DTE features two stable isomers that differ in HOMO energy. In a hole-conducting matrix with a certain HOMO energy, one DTE isomer acts as a hole trap, while the other isomer has a negligible influence on charge transport. The energetic trap depth is varied by blending DTE with various TPD-type hole conductors featuring different HOMO levels. In first proof-of-principle experiments, the hole mobility of the space-charge-limited current in a blend with DTE is reversibly switched by a factor of ca. 20. In contrast, in blends where DTE is replaced by a permanent trap with a similar trap depth trap-limited current is observed. Temperature dependent current density-voltage measurements and impedance spectroscopy are applied.

[1] R.C. Shallcross *et al.*, Adv. Mater. **25**, 469 (2013).

CPP 55.5 Thu 12:00 ZEU 222

**Revisiting the Concept of Carrier Heating in Hopping Transport** — ●MARKUS KRAMMER<sup>1</sup>, CHRIS GROVES<sup>2</sup>, and KARIN ZOJER<sup>1</sup> — <sup>1</sup>Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Austria — <sup>2</sup>School of Engineering and Computing Science, Durham University, United Kingdom

Charges can be viewed to migrate through amorphous materials like disordered organic semiconductors due to hopping between localised states. This motion is governed by a complex interplay of energetic disorder, electric field, interactions, temperature and other parameters. The concept of carrier heating holds the promise of being able to conveniently predict field-dependent steady state charge carrier densities (SSCCD). [1] The SSCCD can be described with a Fermi Dirac distribution such that the impact of an electric field manifests in an effective temperature; provided that electric field and interactions are weak enough. As the conditions for the validity of this concept are still debated [2], we provide here the exact analytical SSCCD for arbitrary field strengths and low charge carrier densities. We demonstrate that the carrier heating effect originates from a modification of the energetic disorder rather than from an effective temperature. By virtue of numerical Kinetic Monte Carlo simulations, the limits of the effective temperature approximation and the field dependence of the effective temperature are revealed.

[1] B. I. Shklovskii *et al.* in Transport, Correlation and Structural Defects, by H. Fritzsche, World Scientific, Singapore (1990), p. 161

[2] S. D. Baranovskii, Phys. Status Solidi B, 251, 487-525 (2014)

CPP 55.6 Thu 12:15 ZEU 222

**On some Fundamentals of Disorder Effects in Organic Semiconductors** — ●FRANK ORTMANN<sup>1</sup>, MICHEL PANHANS<sup>1</sup>, ANDREAS LÜCKE<sup>2</sup>, JOHANNES BENDUHN<sup>3</sup>, SEBASTIAN SCHELLHAMMER<sup>1</sup>, GI-ANAURELIO CUNIBERTI<sup>1</sup>, UWE GERSTMANN<sup>2</sup>, KOEN VANDEWAL<sup>3</sup>, and WOLF-GERO SCHMIDT<sup>2</sup> — <sup>1</sup>Institute for Materials Science, Dresden Center for Computational Material Science and Center for Advancing Electronics Dresden, TU Dresden — <sup>2</sup>Lehrstuhl für Theoretische Materialphysik, Universität Paderborn — <sup>3</sup>Dresden Integrated Center for Applied Physics and Photonic Materials, TU Dresden

Disorder is ubiquitous in Organic Semiconductors but its quantification is rather difficult and depends on the measurement technique. In addition to its magnitude, an open question is if dynamic (thermal) and static disorders behave similar. We show examples of our recent research that shed light on the influence of a static theoretical treatment of vibrational dynamics in transport but also on the possibility of reducing thermal disorder by quantization.

CPP 55.7 Thu 12:30 ZEU 222

**Charge Transport in Rubrene Crystals: Dynamic Disorder and Polarons** — ●MICHEL PANHANS<sup>1</sup>, DESANKA BOSKOVIC<sup>2</sup>, PABLO ORDEJÓN<sup>2,3</sup>, and FRANK ORTMANN<sup>1</sup> — <sup>1</sup>Institute for Material Science, Dresden Center for Computational Material Science and Center for Advancing Electronics Dresden, TU Dresden — <sup>2</sup>ICN2 - Institut Catala de Nanociencia i Nanotecnologia, Barcelona — <sup>3</sup>CSIC - Consejo Superior de Investigaciones Cientificas, Barcelona

The understanding of charge transport in small-molecule organic semiconductors remains a challenge despite various experimental and theoretical studies in recent years. This is mainly because transport phenomena in organic semiconductors are strongly influenced by vibrations and disorder effects (such as traps, dopants, structural disorder, etc.).

We investigate the temperature dependent p-type charge transport in rubrene crystals including the electron-phonon interaction. This interaction can be either described in terms of vibrational disorder or polaronic effects with different implications for the temperature dependent carrier mobility. We present a new mixed approach with an empirical phonon cutoff energy to study the transition from a semiclassical to a quantum description of electron-phonon interaction.

CPP 55.8 Thu 12:45 ZEU 222

**Impact of Polaron Delocalization on Electrical Transport in Conjugated Polymers** — ROBERT STEYRLLEUTHNER, FELIX KRAFFERT, and ●JAN BEHRENDIS — Berlin Joint EPR Lab, Fachbereich Physik, Freie Universität Berlin, Germany

The degree of charge delocalization strongly influences the transport properties of organic semiconductors. However, accurately determining the spatial extension of polaron wave functions remains a challenge. Using electron nuclear double resonance (ENDOR) spectroscopy for measuring the hyperfine coupling between photogenerated polarons and bound nuclear spins, we investigate the delocalization of holes in the semicrystalline conjugated polymer PBTTT (Poly[2,5-bis(3-hexadecylthiophen-2-yl)thieno[3,2-b]thiophene]). An extrapolation of the corresponding oligomer spectra reveals that charges tend to delocalize over 4.0 to 4.8 nm with delocalization strongly dependent on molecular order and crystallinity of the PBTTT polymer thin films. [1]

We further use ENDOR spectroscopy to measure the charge delocalization for a number of conjugated polymers with applications in optoelectronic devices. The results are compared to charge carrier mobilities obtained from time-of-flight measurements, and a correlation between polaron extension and carrier mobility will be discussed.

[1] R. Steyrlleuthner, Y. Zhang, L. Zhang, F. Kraffert, B.P. Cherniawski, R. Bittl, A.L. Briseno, J.-L. Bredas and J. Behrends, Phys. Chem. Chem. Phys., 2016, DOI: 10.1039/c6cp07485e