

CPP 30: Transport: Molecular Electronics and Photonics (jointly with CPP, HL, MA, O)

Time: Wednesday 9:30–12:45

Location: HSZ 201

CPP 30.1 Wed 9:30 HSZ 201

Negative differential conductance in single-molecule junctions with ferromagnetic electrodes — ●PETER HASCH, YUXIANG GONG, LI JIANG, CARLOS-ANDRES PALMA, JOACHIM REICHERT, and JOHANNES V. BARTH — Physics Department E20, Technische Universität München, Germany

Scaling down logic operations to the level of single molecules might be considered the next frontier in computation. One approach is to electrically control single spin states in a molecule, trapped between two electrodes.

Here we report the observance of a negative differential conductance (NDC), measured in a single-molecule junction. The investigated NDC could be explained by a single-spin phenomenon, arising when the molecule gets charged due to voltage-induced depopulation of the highest occupied molecular orbital. This oxidation is monitored by Raman spectroscopy, which allows to analyze chemical and electronic structures with a single-molecule sensitivity. By (anti)ferromagnetic coupling of the unpaired spin on the molecule to one of the electrodes, the molecule might act as a spin-valve, blocking charge transport of the opposite spin direction.

Single-molecule NDC elements bear considerable potential for logical crossbar architectures, and could shrink the actual state of the art devices significantly in size.

CPP 30.2 Wed 9:45 HSZ 201

Single-molecule junctions with oligoynes and epitaxial graphene nanoelectrodes — ●KONRAD ULLMANN¹, SUSANNE LEITHERER², MAXIMILIAN KREMPE³, RIK TYKWIŃSKI³, MICHAEL THOSS², and HEIKO WEBER¹ — ¹Lehrstuhl für Angewandte Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — ²Institut für Theoretische Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — ³Organische Chemie I, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany

Single molecule junctions using graphene as electrode material have drawn considerable attention in recent years [1,2]. Due to their open access architecture, their transparency and robustness, they are well suited for a variety of unprecedented experiments. The electrode material graphene also allows for new anchor groups which connect the molecule to the electrode, e.g. via Pi-interaction.

We report on experiments using an oligoyno molecular wire with platinum termination, being contacted with epitaxial graphene nanoelectrodes. I-V characteristics show a linear behavior with a conductance close to the conductance quantum. Furthermore we present an experimental setup which uses electrospray ionisation to bring the molecules in contact with the graphene electrodes.

[1] K. Ullmann et al., *Nano Lett.* **15**, 3512, 2015[2] F. Prins et al., *Nano Lett.* **11**, 4607, 2011

CPP 30.3 Wed 10:00 HSZ 201

Visualizing the role of molecular orbitals in charge transport through individual diarylethene isomers — ●GAËL REECHT¹, CHRISTIAN LOTZE¹, DMYTRO SYSOIEV², THOMAS HUHN², and KATHARINA J. FRANKE¹ — ¹Freie Universität Berlin, Berlin, Germany — ²Universität Konstanz, Konstanz, Germany.

Diarylethene molecules are prototype molecular switches with their two isomeric forms exhibiting strikingly different conductance, while maintaining similar length. With a scanning tunneling microscope (STM) we investigate the electronic structure and the transport properties of the open and closed isomers of a sulfur-free diarylethene. The electronic structure is determined with scanning tunneling spectroscopy (STS) for the molecule lying on the surface. Between the two isomers, intriguing differences of the energy and the spatial extend of the molecule orbitals are observed. We then lift the two isomers with the tip of the STM and measure the current passing through the individual molecules. We observe an important difference of conductance between the two forms. With a simple analytical model of transport based on the results of the STS measurements, we show that the previously determined orbital characteristics are essential ingredients for the complete understanding of the transport properties.

CPP 30.4 Wed 10:15 HSZ 201

Electronic transport properties of a tripodal molecular plat-

form — ●SAFA GOLROKH BAHOOOSH, AMIN KARIMI, ELKE SCHEER, and FABIAN PAULY — Department of Physics, University of Konstanz, 78457 Konstanz, Germany

Intensive studies on single-molecule junctions have been performed to explore the implementation of molecular-scale devices and to understand how the molecules transport charges[1]. While molecules with delocalized π -systems are the ideal compounds to form wires for electronic applications due to their expected high conductance, tripodal molecular platforms that points almost perpendicular to the surface, appear as promising candidates to establish a conducting path between two electrodes. To take into account these aspects, a 9,9'-spirobifluorene (SBF) platform has been introduced. By combining experimental and theoretical investigations of elastic and inelastic charge transport, we show that the current proceeds through the molecular "backbone" and identify a binding geometry that is compatible with the experimental observations in mechanically controlled break junctions[2]. The conductive molecular wire on the platform features a well-defined and relatively high conductance despite the length of the current path of more than 1.7 nm. If time permits, the possibility to use these molecules as a molecular toggle switch, as observed in subsequent studies with a scanning tunneling microscope, will be discussed.

[1] S. Aradhy, and L. Venkataraman, *Nature Nanotechnol.* **8**, 399 (2013)[2] M. A. Karimi, S. G. Bahoosh, M. Valášek, M. Bürkle, M. Mayor, F. Pauly, and E. Scheer, *Nanoscale* **8**, 10582 (2016)

CPP 30.5 Wed 10:30 HSZ 201

Analysis of local current through molecular wires in open quantum systems — ●DAIJIRO NOZAKI, ANDREAS LÜCKE, and WOLF GERO SCHMIDT — Lehrstuhl für Theoretische Materialphysik, Universität Paderborn, 33095 Paderborn, Germany

The understanding of the local electronic flows in single molecules is of fundamental importance in the design of functional molecules as well as molecule-based electronic devices [1-4]. The charge transport through molecular wires connected between contacts is investigated using non equilibrium Green's function formalism combined with Landauer formula. Energy-dependent and total current through a series of molecular junctions are calculated in real space representation. The influence of contact positions, functional groups, and the replacement of atoms as defects on the transport properties are examined systematically. The static current-induced local magnetic field is also investigated in carbon-based molecular wires. It is shown that even in the same bias directions the direction of the magnetic field is easily reversed depending on the molecular topologies and the positions of electric contacts.

[1] M. Walz, J. Wilhelm, and F. Evers *Phys. Rev. Lett.* **113**, 136602 (2014)[2] J. Wilhelm, M. Walz, and F. Evers *Phys. Rev. B* **92**, 014405 (2015)[3] G. C. Solomon, C. Herrmann, T. Hansen, V. Mujica, and M. A. Ratner *Nat. Chem.* **2**, 223 (2010)[4] T. Ono and K. Hirose, *Phys. Rev. Lett.* **98**, 026804 (2007)

CPP 30.6 Wed 10:45 HSZ 201

Controlling the conductance of graphene-molecule junctions by proton transfer — ●DOMINIK WECKBECKER, PEDRO B. COTO, and MICHAEL THOSS — FAU Erlangen-Nürnberg, Institut für Theoretische Physik, Staudtstrasse 7/B2, 91058 Erlangen, Germany

The possibility of using single-molecule junctions as components of nanoelectronic devices has motivated intensive experimental and theoretical research on the underlying transport mechanism in these systems [1]. In this contribution, we investigate from a theoretical perspective intramolecular proton transfer reactions as a mechanism for controlling the conductance state of graphene-based molecular junctions. Employing a methodology that combines first-principles electronic structure calculations with nonequilibrium Green's function transport theory [1], we show that the proton transfer reaction proceeds via a two-step mechanism and gives rise to several states of the junction with different conductance properties. In addition, we demonstrate that the relative stability of the different conductance states and the energy barriers for the interconversion reactions can be controlled by means of an external electrostatic field. The possibility of using this mechanism for the design of nanomolecular devices such as diodes or switches

is also discussed [2,3].

[1] Cuevas, J. C. and Scheer, E., *Molecular Electronics*, World Scientific Pub. Co., Singapore, 2010

[2] Hofmeister, C. *et al.*, *J. Mol. Model.* **20**, 2163 (2014)

[3] Hofmeister, C. *et al.*, arXiv: 1611.01027v1 (2016)

15 min. break.

CPP 30.7 Wed 11:15 HSZ 201

Hierarchical Quantum Master Equation Approach to Vibrationally Coupled Nonequilibrium Charge Transport in Single-Molecule Junctions — ●CHRISTIAN SCHINABECK¹, ANDRÉ ERPENBECK¹, RAINER HÄRTLE², and MICHAEL THOSS¹ — ¹Institut für Theoretische Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany. — ²Institut für Theoretische Physik, Georg-August-Universität Göttingen, Göttingen, Germany.

We investigate vibrationally coupled transport in single-molecule junctions using the hierarchical quantum master equation (HQME) approach [1-3]. This method allows a systematic convergence of the reduced dynamics of open quantum systems beyond the traditional perturbative master equations. We demonstrate the importance of vibrational nonequilibrium effects for a model molecular junction consisting of an electronic level coupled to fermionic leads as well as a vibrational mode. In particular, in the off-resonant transport regime, the inelastic cotunneling signal is analyzed for a vibrational mode in full nonequilibrium, revealing a complex interplay of different transport processes and deviations from the commonly used $G_0/2$ rule of thumb [3]. Moreover, an extension of the HQME approach is presented, which allows the calculation of the full counting statistics. Using this method, the influence of higher-order cotunneling processes on the current fluctuations is analyzed.

[1] Y. Tanimura *et al.*, *J. Phys. Soc. Jpn.* **75**, 082001 (2006).

[2] J. Jin *et al.*, *J. Chem. Phys.* **128**, 234703 (2008).

[3] C. Schinabeck *et al.*, *Phys. Rev. B* **94**, 201407 (2016).

CPP 30.8 Wed 11:30 HSZ 201

Theoretical study of current-induced bond rupture in molecular junctions — ●ANDRÉ ERPENBECK, CHRISTIAN SCHINABECK, LUKAS GÖTZENDÖRFER, and MICHAEL THOSS — Institut für Theoretische Physik und Interdisziplinäres Zentrum für Molekulare Materialien (ICMM), Friedrich-Alexander-Universität Erlangen-Nürnberg, Staudtstr. 7/B2, D-91058 Erlangen, Germany

Electronic-vibrational coupling in charge transport through single molecule junctions may result in current-induced bond rupture and is thus an important mechanism for the stability of the junction. In this contribution, we demonstrate how the hierarchical quantum master equation (HQME) theory in combination with the quasi-classical Ehrenfest approach for the nuclear degrees of freedom can be used to simulate current-induced bond rupture in single molecule junctions. Employing generic models for molecular junctions with dissociative nuclear potentials, we analyze the underlying mechanisms. In particular, we investigate the dependence of the current, the population and the dissociation probability on the model parameters. In addition, we validate the quasi-classical Ehrenfest approach using numerically exact results obtained by the HQME method [1] for a model comprising one harmonic vibrational mode.

[1] C. Schinabeck, A. Erpenbeck, R. Härtle, M. Thoss, *Phys. Rev. B* **94**, 201407(R) (2016)

CPP 30.9 Wed 11:45 HSZ 201

Spin Transport in Helical Systems — ●MATTHIAS GEYER^{1,2}, RAFAEL GUTIÉRREZ¹, STEFAN SIEGMUND³, and GIANAURELIO CUNIBERTI^{1,2} — ¹Institute for Materials Science, TU Dresden, 01062 Dresden, Germany — ²Dresden Center for Computational Materials Science, TU Dresden, 01062 Dresden, Germany — ³Center for Dynamics, TU Dresden, 01062 Dresden, Germany

Various experiments have shown strong spin selectivity in chiral molecules like DNA at room temperature. Since atomic spin orbit coupling alone is insufficient to explain the effect's magnitude, a relation to the helical geometry has been suggested. We want to provide a better understand of the underlying mechanisms by analytically and numerically investigating suitable models for electrons in helical systems with spin orbit coupling.

We follow two complementary approaches: a generic and simplified model to study the bare influence of the helical geometry and a more realistic one to calculate the effect for specific molecules. The for-

mer starts with a 3D continuum model with helix-shaped confinement from which an effective 1D Hamiltonian is derived using adiabatic perturbation theory. For the latter an effective tight-binding model is derived from the microscopic Hamiltonian of a specific molecule. Incoherent transport calculations are performed for both models using master equations with dephasing, accounting for decoherence due to the coupling to vibrational degrees of freedom arising from structural fluctuation.

CPP 30.10 Wed 12:00 HSZ 201

Charge Carrier Dynamics in κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl: From Mott Insulator to Quantum Spin Liquid — ●JANA-ISABELLE POLZIN¹, BENEDIKT HARTMANN¹, TAKAHIKO SASAKI², and JENS MÜLLER¹ — ¹Institute of Physics, Goethe University Frankfurt, Germany — ²Institute for Materials Research, Tohoku University, Sendai, Japan

The organic charge transfer salts κ -(ET)₂X are model systems for studying strongly-correlated charge carriers and the Mott metal-insulator transition in reduced dimensions. Recently, the influence of quenched disorder attracted considerable attention. Conducting layers of ET molecules are separated by thin, insulating sheets with anions X, resulting in a quasi-2D electronic band structure. Within the ET layers the molecules are arranged in dimers forming a triangular lattice. One free charge carrier exists per dimer, its spin being geometrically frustrated. The Mott insulator κ -(ET)₂Cu[N(CN)₂]Cl shows antiferromagnetic ordering at $T_N \approx 27$ K. It has been shown that increasing disorder induced by X-ray irradiation drives the Mott insulating state with long-ranged antiferromagnetic order into a quantum spin liquid state [1]. Here, we discuss comparative measurements of fluctuation spectroscopy on pristine and irradiated samples in order to investigate the changes in electronic transport mechanism and low-frequency charge carrier dynamics [2] when tuning the Mott insulator to the spin liquid ground state. This results in a decrease of both resistivity and the current or voltage fluctuations after irradiation.

[1] PRL 115, 077001 (2015)

[2] PRL 114, 216403 (2015)

CPP 30.11 Wed 12:15 HSZ 201

Charge Carrier Dynamics at the Mott transition in κ -(BEDT-TTF)₂Cu[N(CN)₂]Br — ●TATJANA THOMAS¹, BENEDIKT HARTMANN¹, TAKAHIKO SASAKI², and JENS MÜLLER¹ — ¹Institute of Physics, Goethe University Frankfurt, Germany — ²Institute for Materials Research, Tohoku University, Sendai, Japan

The organic charge transfer salts κ -(ET)₂X are considered as model systems for studying the Mott metal-insulator transition – a key phenomenon in the physics of strongly correlated electrons – in reduced dimensions. In particular, the influence of disorder on the criticality of the Mott transition recently has been a matter of debate. Partially deuterated κ -[(H₈-ET)_{0.2}(D₈-ET)_{0.8}]₂Cu[N(CN)₂]Br, which is located in the critical region of the phase diagram, can be fine-tuned through the Mott transition by utilizing a glass-like structural ordering transition of the ET molecules' terminal ethylene groups. By applying different thermal relaxation protocols, both the ratio of W/U and a small degree of quenched disorder can be set at will, the former corresponding to changes in hydrostatic pressure of ~ 200 bar. We employ fluctuation (noise) spectroscopy as a powerful tool to study the charge carrier dynamics at low frequencies. When crossing the S-shaped Mott transition line, surprisingly we observe a step-like increase of the resistance fluctuations in the metallic region. We discuss our results in terms of critical slowing down of the order parameter fluctuations [1] and electronic phase separation, and an extended region of the phase diagram where the fluctuations are non-Gaussian.

[1] B. Hartmann *et al.*, *Phys. Rev. Lett.* **114**, 216403 (2015).

CPP 30.12 Wed 12:30 HSZ 201

Thermal conductance of Teflon and Polyethylene: Insight from an atomistic, single-molecule level — ●MARIUS BUERKLE and YOSHIHIRO ASAI — AIST, Tsukuba, Japan

The thermal transport properties of teflon (polytetrafluoroethylene) and its polyethylene counterparts are, while highly desirable and widely used, only superficially understood. Here, we aim therefore to provide rigorous insight from an atomistic point of view in context of single-molecule devices. We show that for vinyl polymers adsorbed on metal-surfaces the thermal transport strongly depends on the properties of the metal-molecule interface and that the reduced thermal conductance observed for teflon derivatives originates in a reduced phonon injection life time. In asymmetric molecules phonon blocking on the

intra molecular interface leads to a further reduction of thermal conductance. For hetrojunctions with different electrode materials we find that thermal conductance is suppressed due to a reduced overlap of the available phonon modes in the different electrodes. A detailed atom-

istic picture is thereby provided by studying the transport through perfluorooctane and octane on a single-molecule level using first principles transport calculations and nonequilibrium molecular dynamic simulations.