TT 65: Transport: Molecular Electronics and Photonics 1
(Joint session of CPP, DS, HL, MA, O and TT organized by TT)

Time: Thursday 9:30–13:00   Location: H23

TT 65.1   Thu 9:30   H23

Pulling and Stretching a Molecular Wire to Tune its Conductance — •Gaël Reecht[1,4], Hervé Bulou[1], Fabrice Scheurer[1], Virginie Speisser[1], Fabrice Mathevet[2], César González[3], Yannick J. Dappe[3], and Guillaume Schull[1] — [1]IPCMS, Strasbourg, France — [2]Laboratoire de Chimie des Polymères, Paris, France — [3]CEA IRAMIS, Saclay, France — [4]Freie Universität Berlin, Berlin, Germany

Molecular junctions are perceived as the ultimate step toward the miniaturization of electronic components based on organic materials. Here, a low temperature scanning tunnelling microscope is used to lift a polythiophene wire from a Au(111) surface while measuring the current traversing the molecular junction. Conductance traces recorded during the lifting procedures reveal abrupt increases of the current intensity, which we associate to detachments of the wire subunits from the surface, in apparent contradiction with the expected exponential decrease of the conductance with wire length. With, ab initio simulations we reproduce the experimental data and demonstrate that this unexpected behavior is due to release of mechanical stress in the wire. Therefore, with the high control ability of the STM, by stretching the suspended molecular wire, we are able to tune its conductance properties.

TT 65.2   Thu 9:45   H23

STM-induced luminescence of single molecule junction — •Michael Chong[1], Gael Reecht[1], Hervé Bulou[1], Alex Boeglin[1], Fabrice Mathevet[2], Fabrice Scheurer[1], and Guillaume Schull[1] — [1]Institut de Physique et Chimie des Matériaux de Strasbourg - CNRS - France — [2]Laboratoire de Chimie des Polymères - CNRS - Université Pierre et Marie Curie, Paris, France

Electroluminescence of a single molecule can be induced by means of scanning tunneling microscopy. When a molecule is placed between two metallic electrodes it is necessary to decouple it using thin insulating layers in order to measure its intrinsic luminescence. A direct contact with the electrodes (tip and substrate), necessary if we envision to build single molecule electronic devices, results in quenching or broadening of the fluorescence of the molecule. We use on-surface polymerization to embed a cromphore molecule in a molecular chain. The STM tip is then used to lift the chain in order to decouple the cromophore from the surface yet mantaining a circuit like configuration trough the molecular chain. The current generated by applying a bias to the electrodes excites the cromophore that then exhibits narrow line luminescence and vibronic peaks allowing chemical identification the emitting unit.

Moreover we demonstrate that this configuration allows to control the lifetime of the excited state of the emitting molecule by two orders of magnitude by changing the coupling of the single molecle with the substrate adjusting the tip-sample separation. This system might open the way to electro-plasmonic devices at the single molecule level.

TT 65.3   Thu 10:00   H23

Effects of spin-orbit coupling and many-body correlations in STM transport through copper phthalocyanine — Benjamin Siegert, •Andrea Donarini, and Milena Grifoni — Institut für Theoretische Physik, Universität Regensburg, 93040 Regensburg, Germany

The interplay of exchange correlations and spin-orbit interaction (SOI) on the many-body spectrum of a copper phtalocyanine (CuPc) molecule and their signatures in transport are investigated. We first derive a minimal model Hamiltonian in a basis of frontier orbitals which is able to reproduce experimentally observed singlet-triplet splittings; in a second step SOI effects are included perturbatively. Major consequences of the SOI are the splitting of former degenerate levels and a sizable magnetic anisotropy, which can be captured by an effective low-energy spin Hamiltonian. We show that STM-based magnetoconductance measurements can yield clear signatures of both these SOI induced effects.

TT 65.4   Thu 10:15   H23

Conductance trend in linear oligoacenes controlled by quantum size-effects — •Richard Korytar[1], Tamar Yelin[2], Nirit Sukenik[2], Ran Vardimon[2], Bharat Kumar[3], Colin Nuckolls[3], Oren Tal[2], and Ferdinand Evers[1] — [1]Institute of Theoretical Physics, University of Regensburg, Germany — [2]Chemical Physics Department, Weizmann Institute of Science, Rehovot, Israel — [3]Department of Chemistry, Columbia University, New York, United States

In conventional electronics, the conductance of a wire decreases with length according to Ohm's law. In molecular electronics, quantum effects lead to a richer phenomenology. Oligoacenes are organic molecules which consist of (linearly) fused benzene rings. Recently, Yelin et al. [submitted] studied conductance of oligoacenes directly coupled to Ag leads and found increase of conductance with molecular length.

I will show that transport through oligoacenes is governed by a quantum size effect which controls the alignment and width of the lowest unoccupied molecular orbital. These ideas will be supported by first-principles transport calculations using density-functional theory.

Linear oligoacenes are one of the simplest realizations of zig-zag terminated graphene nano-ribbons. In the long-wire limit, I will demonstrate that the conductance as a function of the molecular length shows surprising oscillations with period of approx. 11 rings [1].

[1] R. Korytár, D. Xenioti, P. Schmitteckert, M. Alouani, and F. Evers, Nature Communications **5**, 5000 (2014).

TT 65.5   Thu 10:30   H23

Investigation of charge transfer processes in single crystals based on π-conjugated molecules — •Antonia Morherr[1], Alisa Chernenkaya[2], Sebastian Witt[1], Katerina Medjanik[3], Michael Bolte[1], Martin Baumgarten[4], Harald O. Jeschke[1], Roser Valentí[1], and Cornelius Krellner[1] — [1]Goethe-Universität Frankfurt, 60438 Frankfurt a. M., Germany — [2]Johannes Gutenberg-Universität, 55099 Mainz, Germany — [3]Lund University, MAX-lab, 22100 Lund, Sweden — [4]MPI für Polymerforschung, 55021 Mainz, Germany

Designing new charge transfer (CT) materials for tuning the physical properties ranging from metallicity over superconductivity to Mott insulators and the understanding of mechanisms of CT is of great interest [1]. New CT crystals of π-conjugated molecules as donors can be obtained by physical vapor transport (PVT) [2]. (Fluorinated) tetracyanoquinodimethane (TCNQ-F$_x$, x=0, 2, 4) was used as acceptor material to grow different CT salts. The crystal structure was detected by X-ray diffraction. Further spectroscopic measurements as infrared and NEXAFS measurements were applied on these single crystals to investigate the CT process. The analysis of N1s and F1s K-edge spectra shows changes for different acceptor strengths. Ab initio calculations for all compounds underline these results. This systematic investigation of CT materials helps to understand the CT process in more detail.

[1] N. Toyota, M. Lang, J. Müller, Low-Dimensional Molecular Metals, Springer-Verlag, Berlin, 2007

[2] B. Mahns et al., Cryst. Growth Des. **14**, 1338 (2014)

TT 65.6   Thu 10:45   H23

Single Molecule Junctions with Epitaxial Monolayer Graphene Electrodes — •Konrad Ullmann[1], Pedro B. Coto[2], Susanne Leitherer[2], Michael Thoss[2], and Heiko B. Weber[1] — [1]Lehrstuhl für Angewandte Physik und Interdisziplinäres Zentrum für Molekulare Materialien, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU) — [2]Institut für Theoretische Physik und Interdisziplinäres Zentrum für Molekulare Materialien, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU)

To study transport through single molecules, a two dimensional, open-access testbed for individual molecules is desirable. Therefore we use epitaxial monolayer graphene to fabricate electrodes for single molecule junctions. With the help of a feedback-controlled electro-burning process nanometer sized gaps can be formed reproducibly. Using these electrodes, we studied transport through molecules with different anchor groups at low temperatures. Strong similarities in results obtained with the MCBJ-technique underline the high quality of our experimental data. For a fullerene-endcapped molecule we are able to assign features from the I-V characteristics to internal molecular degrees of

freedom [1].
[1] K. Ullmann et al., Nano Lett. **15**, 3512 (2015)

TT 65.7　Thu 11:00　H23
Simulation of Electron Transport through Graphene-Molecule Junctions — •Susanne Leitherer[1], Uwe Frank[1], Konrad Ullmann[2], Pedro B. Coto[1], Heiko Weber[2], and Michael Thoss[1] — [1]Institute for Theoretical Physics and Interdisciplinary Center for Molecular Materials, University Erlangen-Nürnberg — [2]Chair of Applied Physics and Interdisciplinary Center for Molecular Materials, University Erlangen-Nürnberg

Charge transport in single-molecule junctions with graphene electrodes is investigated using a combination of density functional theory (DFT) electronic structure calculations and Landauer transport theory. In particular, we study covalently bonded molecule-graphene junctions as well as junctions, where the molecule is weakly bonded to graphene by van der Waals interaction [1]. Considering different examples for molecular bridges between graphene electrodes, we analyze the transmission probability and current-voltage characteristics. In junctions with zigzag terminated graphene electrodes, we find edge states, which can induce additional transport channels [2]. Furthermore, local conductance properties are investigated in the nanojunctions.
[1] K. Ullmann et al., Nano Lett. **15**, 3512 (2015)
[2] I. Pshenichnyuk et al., J. Phys. Chem. Lett. **5**, 809 (2013)

15 min. break

TT 65.8　Thu 11:30　H23
Electron transport through C_{20} molecular junction — •Shigeru Tsukamoto and Stefan Blügel — PGI-1/IAS-1, Forschungszentrum Jülich and JARA, Jülich, Germany

We present electron transport properties of C_{20} molecular junctions, which are evaluated within the framework of the density functional theory. The C_{20} molecular junctions employed in this work are composed of a pair of Al bulk electrodes and a single C_{20} molecule, which is known as the smallest fullerene molecule. The scattering wave functions of the molecular junctions are calculated by solving the Kohn-Sham equation by means of the over-bridging boundary matching method, which is based on the real-space finite-difference formalism. The transmission properties are extracted from the scattering wave functions and the electron transmissions are evaluated by the Landauer-Büttiker formula. The electron transmissions and the scattering wave functions are further analyzed by using the eigenchannel decomposition technique. As the result of the eigenchannel analysis, although the total transmission value is $\sim 3.0G_0$ at around the Fermi level, more than five transmission channels are found to contribute to the electron transport, and none of the eigenchannels are opened to 100%. From the spatial distributions of the eigenchannels, we can see that the HOMO states of C_{20} molecule, which are three-fold degenerated and occupied to one-third, mainly contribute to the transport. In addition, the LUMO state is also found to contribute as one of the eigenchannels at around the Fermi level. In the talk, we will present electron transport calculations with different molecular orientations.

TT 65.9　Thu 11:45　H23
Quantum interference effect transistor via "Kondo Blockade" in single molecule junctions — •Andrew Mitchell[1] and Jens Paaske[2] — [1]Institute for Theoretical Physics, Utrecht University, 3584 CE Utrecht, The Netherlands — [2]Niels Bohr Institute, University of Copenhagen, DK-2100 Copenhagen, Denmark

Single molecule junctions are basic building blocks of molecular electronics devices. The full power of these devices will be realized by exploiting inherent quantum mechanical effects. Two of the most striking quantum phenomena, with no classical analogue, are quantum interference (QI) due to competing electron transport pathways, and the Kondo effect (KE) due to entanglement and strong electronic interactions. Both QI and KE are widely observed in experiments. The description of QI accounts for the complexities of molecular structure, but is typically non-interacting. By contrast, the Anderson impurity model is usually used to describe interactions and the Kondo effect, but totally neglects molecular structure. In this talk I discuss the subtle interplay between QI and KE in a unified theory, showing that a novel gate-tunable "Kondo Blockade" regime can be exploited to realize an efficient quantum interference effect transistor.

TT 65.10　Thu 12:00　H23
Quantitative *ab initio* simulations of nanocarbon-metal extended contacts — •Artem Fediai[1,2], Dmitry Ryndyk[1,2], and Gianaurelio Cuniberti[1,2,3] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden — [2]Center for Advancing Electronics Dresden, TU Dresden — [3]Dresden Center for Computational Materials Science, TU Dresden, 01062 Dresden, Germany

Recently developed approach presented in [1] allows to get quantitative information about the resistance R_c, effective contact length L_c, and contacts resistance scaling $R_c(L_c)$ in different extended side contacts depending on the electrode material. We apply this approach to find a contact resistance of side CNT-metal contacts, transfer length in graphene-metal contacts and electronic properties of the diodes with CNT channel and asymmetric contacts (with the electrodes made of different metals). These kinds of *ab initio* simulations were previously impossible due to numerical intractability of the side contacts longer then several nanometers. Our approach explicitly uses extended contact model concept, enforced by modular approach. This allows us to overcome numerical problems and understand physical processes in extended contacts.
[1] A. Fediai, D.A. Ryndyk, G. Cuniberti, PRB **91**, 165404 (2015)

TT 65.11　Thu 12:15　H23
Molecular switches for dangling bond circuits — •Thomas Lehmann[1,2], Dmitry A. Ryndyk[1,2], and Gianaurelio Cuniberti[1,2] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, Germany — [2]Dresden Center for Computational Materials Science (DCMS), TU Dresden, Germany

On the road to atomic-scale electronic circuits, dangling bond wires are promising candidates. Dangling bonds are formed by selectively removing hydrogen from a passivated silicon surface [1,2] and multiple dangling bonds in a row feature extended electronic states. Those quasi 1D surface structures can be used as atomic scale interconnects. In such circuits, molecules, which can controllably passivate or depassivate a dangling bond can provide logical inputs for constructing simple logic elements. In this talk, we present recent studies combining density-functional based approaches with Green function methods of a molecular switch for dangling bond wires on silicon.
[1] T. Hitosugi, T. Hashizume, S. Heike, S. Watanabe, Y.Wada,
　　T. Hasegawa, K. Kitazawa, Jpn. J. Appl. Phys. **36**, L361 (1997)
[2] H. Kawai, F. Ample, Q. Wang, Y. K. Yeo, M. Saeys, C. Joachim,
　　J. Phys. Condens. Matter **24**, 095011 (2012)

TT 65.12　Thu 12:30　H23
Switchable negative differential resistance induced by quantum interference effects in porphyrin-based molecular junctions — •Daijiro Nozaki[1], Lokamani Lokamani[2], Alejandro Santana-Bonilla[2], Arezoo Dianat[2], Rafael Gutierrez[2], Gianaurelio Cuniberti[2], and Wolf Gero Schmidt[1] — [1]Lehrstuhl für Theoretische Physik, Universität Paderborn, Paderborn, Germany — [2]Institute for Materials Science, TU Dresden, Dresden, Germany

Charge transport through a carbon-based molecular switch consisting of different tautomers of metal-free porphyrin embedded between graphene nanoribbons is studied by combining electronic structure calculations and nonequilibrium Green's function formalism. Different low-energy and low-bias features are revealed, including negative differential resistance (NDR) and antiresonances, both mediated by subtle quantum interference effects. Moreover, the molecular junctions can display moderate rectifying or nonlinear behavior depending on the position of the hydrogen atoms within the porphyrin core. We rationalize the mechanism leading to NDR and antiresonances by providing a detailed analysis of transmission pathways and frontier molecular orbital distribution.
[1] D. Nozaki, J. Phys. Chem. Lett. **6**, 3950 (2015).

TT 65.13　Thu 12:45　H23
Base alignment dependence on Seebeck coefficient of DNA: A diagrammatic non-equilibrium transport theory approach — •Yoshihiro Asai[1], Yueqi Li[2], Limin Xiang[2], Julio L. Palma[2], and Nongjian Tao[2] — [1]Research Center for Computational Design of Advanced Functional Materials, AIST, Central 2, Umezono 1-1-1, Tsukuba, Ibaraki 305-8568, Japan — [2]Center for Bioelectronics and Biosensors, Biodesign Institute, Arizona State University, Tempe, Arizona 85287-5801, USA

Theoretical calculation of temperature dependence of transport properties at finite bias voltage and/or at finite temperature gradient re-

quires careful description of low energy excitations. Incorporation of phonon transport and its coupling to electron transport by no means should play a crucial role to describe the low energy physics. One of the authors succeeded to describe theoretically the temperature cross over behavior of the electric conductance found in the experiment of a long oligothiophene single molecular wires. The diagrammatic non-equilibrium transport theory is useful to describe the problem qualitatively. While the necessity of the non-perturbative approach to the problem is clear, it would be interesting to know how far we could go within the perturbative framework given that any reliable non-perturbative approach for the problem is not available at present. Here, we apply the theory to discuss the base alignment dependence of the Seebeck coefficient of DNA in the hopping temperature region. We will make comparative discussions on our theoretical results with our experimental ones.