

TT 32: Transport: Nanoelectronics III - Molecular Electronics 1

Time: Wednesday 15:00–17:30

Location: BH 334

TT 32.1 Wed 15:00 BH 334

Influence of disorder and dephasing events on the electron transport through conjugated molecular wires in molecular junctions — ●DAIJIRO NOZAKI¹, CLAUDIA GOMES DA ROCHA¹, HORACIO M. PASTAWSKI², and GIANAURELIO CUNIBERTI¹ — ¹Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany — ²Instituto de Física Enrique Gaviola (CONICET) and FaMAF, Universidad Nacional de Córdoba, Argentina

Understanding electron transport mechanisms on the molecular scale is a central issue in the field of molecular electronics. Among them, the dephasing effect, causing the tunneling-to-hopping transition, has a great importance for applications as well as from the fundamental point of view. In this study, we analyzed the coherent and incoherent electron transmission through conjugated molecular wires by means of a first principle approach within D'Amato-Pastawski model [1]. Our approach can study explicitly the structure/transport relationship in molecular junctions in a dephasing environment using only a single tuning parameter. We investigated the length dependence and the influence of thermal disorder on the transport and reproduced the well-known tunneling-to-hopping transition [2]. This approach will be a powerful tool for the interpretation of recent conductance measurements of molecular wires.

[1] J. L. D'Amato and H. M. Pastawski, *Phys. Rev. B* 41, 7411 (1990).

[2] D. Nozaki, C. G. Rocha, H. M. Pastawski, and G. Cuniberti, submitted.

TT 32.2 Wed 15:15 BH 334

Master-equation approach for simulating STM images of single molecules — ●TIM LUDWIG¹, CARSTEN TIMM¹, TORSTEN HAHN², and JENS KORTUS² — ¹TU Dresden, Dresden, Germany — ²TU Freiberg, Freiberg, Germany

The power of STM imaging of single molecules lies in the spatial resolution and a well-defined contact with highly controllable contact strength. Therefore a wide range of molecule-substrate combinations are investigated by this technique. Traditional methods for simulating STM images, e.g., DFT + Tersoff-Hamann, use a static single-particle density-of-states description of the scanned object. This can be inappropriate for the investigation of single molecules, as for example Coulomb blockade may occur. In contrast a master-equation approach models the full many-body dynamics on the molecule, including, e.g., spin blockade and vibrational coupling. Furthermore it can be used for arbitrarily high bias voltages. We report on the first attempt for simulating STM images of single molecules by using a master-equation description.

TT 32.3 Wed 15:30 BH 334

Molecular orbital switching in photochromic single molecular junctions — ●YOUNGSANG KIM¹, TORSTEN PIETSCH¹, ELKE SCHEER¹, THOMAS HELLMUTH², FABIAN PAULY², DMITRO SYSOIEV³, THOMAS HUHN³, THOMAS EXNER³, ULRICH GROTH³, ULRICH STEINER³, and ARTUR ERBE⁴ — ¹Department of Physics, Universität Konstanz, 78467 Konstanz, Germany — ²Institut für Theoretische Festkörperphysik, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany — ³Department of Chemistry, Universität Konstanz, 78467 Konstanz, Germany — ⁴Helmholtz-Zentrum Dresden Rossendorf, 01328 Dresden, Germany

Photoswitchable molecules gain significant interest due to the applicability in data storage media, as optical switches, and in novel logic circuits [1]. The molecular energy levels and their coupling to the metal electrodes determine both the single-molecule conductance and the switching ratio between the two states of a switch molecule. Here, we show the preferential conductance of individual, specifically designed sulfur-free diarylethene [2] molecules bridging mechanically controlled break-junction (MCBJ) electrodes at low temperatures. These molecules undergo a ring-opening/ring closure transition upon irradiation with light of suitable wave-length. We discuss the changes of molecular energy levels and electrode couplings in both isomers obtained by evaluating the current-voltage (I-V) characteristics using the single-level model [3].

[1] M. Del Valle et al., *Nat Nanotechnol* 2, 176 (2007).

[2] D. Sysoiev et al., *Chem. Eur. J.* 17, 6663 (2011).

[3] Y. Kim et al., *Nano Lett.* 11, 3734 (2011); L. Zotti et al., *Small* 6, 1529 (2010).

TT 32.4 Wed 15:45 BH 334

Vibrationally dependent electron-electron interactions in single-molecule junctions: A mechanism for asymmetric gate-voltage dependence of the current in symmetrically coupled junctions — ●RAINER HÄRTLE, ANDRE ERPENBECK, and MICHAEL THOSS — Institut für Theoretische Physik, Friedrich-Alexander-Universität, Erlangen-Nürnberg, Staudtstr. 7/B2, D-91058, Germany

The influence of non-adiabatic effects due to vibrationally dependent electron-electron interactions on the transport characteristics of single-molecule junctions is investigated. Non-adiabatic effects in single-molecule junctions result from the dependence of the electronic states of the molecular bridge on the nuclear coordinates. This includes a direct coupling between electronic states, which is important, for example, in the presence of avoided level-crossings or conical intersections, but involves also vibrationally dependent electron-electron interactions. We demonstrate that the latter gives rise to an effective electronic-vibrational coupling, which depends on the population of the electronic states. As a result, the current-voltage characteristics of a single-molecule junction can be asymmetric with respect to a gate voltage, even if the molecular bridge is symmetrically coupled to the leads. To describe these effects, we employ a master equation approach that is based on a second-order expansion in the coupling between the molecule and the leads.

TT 32.5 Wed 16:00 BH 334

Transport through π -stacked benzene rings and evaluation of their IETS — ●THOMAS HELLMUTH, MARIUS BÜRKLE, FABIAN PAULY, and GERD SCHÖN — Institut für Theoretische Festkörperphysik, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany

Motivated by recent experiments [1], we study the electric transport through wires with multiple π - π -stacked benzene rings connected to gold electrodes. In our density-functional-theory-based calculations we analyze various junction geometries and study both the elastic transmission and inelastic electron tunneling spectra (IETS). Transmission eigenchannels show that the current is mostly carried by the π -system, but the gold-benzene binding affects the transmission path notably. Features in the inelastic tunneling spectra are identified with their corresponding vibrational modes.

[1] S. T. Schneebeli et al., *J. Am. Chem. Soc.* 133, 2136 (2011)

15 min. break.

TT 32.6 Wed 16:30 BH 334

Coulomb blockade in molecular junctions with intermediate coupling to the leads — ●DMITRY RYNDYK, ANDREA DONARINI, MILENA GRIFONI, and KLAUS RICHTER — Institute for Theoretical Physics, University of Regensburg, 93040 Regensburg, Germany

An ab initio based theoretical approach to describe nonequilibrium many-body effects in molecular transport is developed. In particular, an Anderson-Hubbard cluster model on the basis of localized molecular orbitals is derived for single-molecule junctions with intermediate coupling to the leads. As an example we consider the Coulomb blockade in a benzenedithiol junction with gold electrodes. An effective few-level model is obtained, transformed into the many-body eigenstate basis and solved within the master equation approach. It is shown that transport through asymmetrically coupled molecular edge states results in strongly suppressed peaks of the differential conductance at small voltage.

TT 32.7 Wed 16:45 BH 334

Spin-polarized transport through single molecules: insight from theory — ●ALEXEI BAGRETS^{1,2} and FERDINAND EVERS^{2,3} — ¹Steinbuch Center for Computing, Karlsruher Institut für Technologie (KIT), Germany — ²Institut für Nanotechnologie, KIT, Germany — ³Institut für Theorie der Kondensierten Materie, KIT, Germany

In this talk, a short overview will be given on our recent advances in DFT modeling of spin-polarized electron transport across single molecules bridged between magnetic reservoirs. In particular, we will discuss implementation of the non-equilibrium Green's function for-

malism, where efficient absorbing boundary conditions encoded into self-energy function are extended to account for the spin-polarized electronic structure of magnetic electrodes. We will demonstrate an ability, within our approach, to treat a variety of different magnetic configurations (e.g. nano-scale domain walls) which could appear when a molecule with few spin-centers (d-ions) is brought in contact with a magnetic surface and a tip of the spin-polarized-STM. Recent experiments [1,2] on spin-transport and magnetoresistance effect observed in hydrogen-phtalocyanine STM molecular junctions will be discussed in detail.

[1] S. Schmaus, A. Bagrets, Y. Nahas, T. K. Yamada, A. Bork, M. Bowen, E. Beaurepaire, F. Evers, W. Wulfhekel, *Nature Nanotechnology* **6**, 185 (2011).

[2] A. Bagrets, A. Jaafar, S. Schmaus, M. Alouani, W. Wulfhekel, F. Evers, preprint (2011)

TT 32.8 Wed 17:00 BH 334

Optical response of a single-molecule junction: current switching, negative differential resistance and plasmon induced conductance enhancement — ●YAROSLAV ZELINSKY^{1,2} and VOLKHARD MAY¹ — ¹Institut für Physik, Humboldt Universität zu Berlin, Newtonstrasse 15, D-12489 Berlin, Germany — ²Bogolubov Institute for theoretical physics National Academy of Science of Ukraine, Metrologichna str. 14b, UA-03683, Kiev, Ukraine

Based on a generalized master equation approach the conductive properties of a current-carrying molecular junction driven by an external optical excitation are studied [1]. The description incorporates vibrational states of the molecule and related relaxation processes with the latter being crucial for the strength of the actual current. A photoinduced switch of the current, the appearance of a negative differential resistance and its photoinduced suppression are predicted [2]. The

nano-electrode plasmon control of the single molecule conductivity appearing at the simultaneous optical excitation of the molecule and the electrodes is also considered [3]. A remarkable enhancement of the current is achieved in this case.

[1] L. Wang, V. May, *Phys.Chem.Chem.Phys.*, **13**, 8755 (2011).

[2] Ya.R. Zelinsky, E.G. Petrov, *Ukr.J.Phys.* **54**, 707 (2009).

[3] Y. Zelinsky, V. May, *Nano Lett.* (submitted).

TT 32.9 Wed 17:15 BH 334

Resonant photoconductance of molecular junctions formed in Gold nano-particle arrays — M. MANGOLD¹, ●J. SCHOPKA¹, M. CALAME², M. MAYOR³, and A.W. HOLLEITNER¹ — ¹Walter Schottky Institut and Physik-Department, Technische Universität München, 85748 Garching — ²Departement Physik, Universität Basel, 4056 Basel, Switzerland — ³Departement Chemie, Universität Basel, 4056 Basel, Switzerland

We report on a molecular phototransistor effect based on oligo(phenylene vinylene) (OPV) incorporated in gold nanoparticle arrays. We find a pronounced photoconductance arising upon resonant excitation of the OPV molecules. We determine the typical response time and the irradiation intensity dependence of this resonant photoconductance. Our results suggest that the charge carriers, which are resonantly excited in the OPV molecules, directly contribute to the current flow through the nanoparticle arrays. We can consistently model the dynamics of the resonant photoconductance by considering the filling and recombination of trap states in the nanoparticle arrays. Our results verify that individual molecules in metal-molecule-metal junctions can be functional modules of optoelectronic devices [1].

[1] M. Mangold, M. Calame, M. Mayor, A.W. Holleitner, *J. Am. Chem. Soc.* **133**, 12185-12191 (2011).