

TT 35: TR: Nanoelectronics III: Molecular Electronics 1

Time: Thursday 14:00–17:45

Location: H21

TT 35.1 Thu 14:00 H21

On-chip optoelectronic functionalization of CNTs with photosynthetic proteins and CdTe nanocrystals — ●FREDERIK HETSCH¹, SIMONE M. KANIBER¹, MATTHIAS BRANDSTETTER¹, FRITZ C. SIMMEL², ITAI CARMELI³, and ALEXANDER W. HOLLEITNER¹ — ¹Walter Schottky Institut, Technische Universität München, Am Coulombwall 3, D-85748 Garching, Germany — ²Physik Department, Technische Universität München, James Franck Straße 1, D-85748 Garching, Germany — ³Department of Chemistry and Biochemistry, Tel-Aviv University, 69978 Tel-Aviv, Israel

We examine carbon nanotube (CNT) based hybrid systems by optoelectronic transport spectroscopy. CNTs are functionalized on-chip both with the "photosystem I" (PSI) and CdTe nanocrystals according to different chemical routes. PSI is a protein complex located in the thylacoid membrane of plants, algae and cyanobacteria which mediates the light-induced electron transfer in the photosynthetic pathway. The efficient energy conversion in the PSI and the adjustable optical properties of colloidal semiconductor nanocrystals, together with a nano-sized dimension, make both systems to promising candidates for applications in nano-optoelectronic devices. We will discuss the optoelectronic properties of PSI-CNT and CdTe-CNT hybrids.

TT 35.2 Thu 14:15 H21

Exact master equation for nano-systems: applications to charge transport through DNA oligomers — ●MATISSE WEI-YUAN TU, MYEONG LEE, STANISLAV AVDOSHENKO, DAIJIRO NOZAKI, RAFAEL GUTIERREZ, and GIANAURELIO CUNIBERTI — Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, 01062 Dresden, Germany

We present a formally exact master equation derived with the help of the Feynmann-Vernon's influence functional theory and a time-dependent electrical current formula derived from it [1,2]. We study charge transport through a DNA 15-mer connected to electrodes, when a single base mismatch is introduced into it. The electronic structure of the molecule is mapped onto an effective linear tight-binding chain with time-fluctuating electronic parameters drawn from snapshots along molecular dynamics trajectories. By exploring the real-time charge propagation and the dynamical current, we analyse the statistical properties of the current for matched and mismatched DNA oligomers.

[1] Matisse Wei-Yuan Tu and Wei-Min Zhang, *Phys. Rev. B* **78**, 235311(2008).

[2] Jinshuang Jin, Matisse Wei-Yuan Tu, Wei-Min Zhang and YiJin Yan, arXiv:0910.1675.

TT 35.3 Thu 14:30 H21

Charge transfer characteristics of dsDNA with base mismatch in a solvent — ●MYEONG H LEE, STANISLAV M AVDOSHENKO, DAIJIRO NOZAKI, RAFAEL GUTIERREZ, and GIANAURELIO CUNIBERTI — Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, 01062, Dresden, Germany

We present charge transfer characteristics of DNA by evaluating the electronic parameters in the presence of water and counterions. Motivated by the recent experiment by Guo *et al.* [1], we investigate three types of dsDNA oligomers; a well matched 15 base pair sequence and single mismatched base pairs (GT, CA) in sequence. Charge transfer parameters are obtained using a fragment orbital approach [2] on the coarse-grained level, where a fragment consists of a pair of nucleotides from each DNA strand. In order to fully take into account the effect of dynamical fluctuations [3] the electronic structure calculation is performed using a hybrid QM/MM method for each snapshot along the MD trajectory. From the electronic structure parameters the current is obtained for both coherent and incoherent transport [4] contribution to see the effect of single base pair mismatch.

[1] X. Guo *et al.*, *Nature Nanotech.* **3**, 163 (2008).

[2] D. G. Fedorov *et al.*, *J. Phys. Chem. A* **111**, 6904 (2007).

[3] R. Gutierrez, *et al.*, *Phys. Rev. Lett.* **102**, 208102 (2009); P. B. Woiczikowski *et al.*, *J. Chem. Phys.* **130**, 215104 (2009); P. B. Woiczikowski *et al.*, *J. Phys. Chem. B* **112**, 7937 (2008).

[4] J. L. D'Amato *et al.*, *Phys. Rev. B* **41**, 7411 (1990).

TT 35.4 Thu 14:45 H21

Conductance Through a Redox System in the Coulomb Blockade Regime: Many-Particle Effects and Influence of Electronic Correlations — ●SABINE TORNOW and GERTRUD ZWICKNAGL — Institut fuer Mathematische Physik, TU Braunschweig, 38106 Braunschweig, Germany

We investigate the transport characteristics of a redox system weakly coupled to leads in the Coulomb blockade regime. The redox system comprises a donor and acceptor separated by an insulating bridge in a solution. It is modeled by a two-site extended Hubbard model which includes on-site and inter-site Coulomb interactions and the coupling to a bosonic bath. The current voltage characteristics is calculated at high temperatures using a rate equation approach. For high voltages exceeding the Coulomb repulsion at the donor site the calculated transport characteristics exhibit pronounced deviations from the behavior expected from single-electron transport. Depending on the relative sizes of the effective on-site and inter-site Coulomb interactions on one side and the reorganization energy on the other side we find negative differential resistance or current enhancement.

TT 35.5 Thu 15:00 H21

Ab-initio based modeling of electron-vibron effects in single-molecule junctions — ●DMITRY A. RYNDYK and KLAUS RICHTER — Institute for Theoretical Physics, University of Regensburg, Germany

We develop an approach for quantum transport in single-molecule junctions, combining an ab initio based many-body model and the techniques of nonequilibrium Green functions. In particular, we use DFT to determine relaxed molecular junction geometry and relevant vibronic modes, and semi-empirical or ab initio (Hartree-Fock based) atomistic calculations for relevant electronic states and the coefficients of electron-vibron interaction. Finally, this effective model is used for transport calculations. As an example, we apply the method to Pt-H₂(O)-Pt junctions and Pt-oligophenyl-Pt junctions. We focus on the interplay of electron-vibron interaction and coupling to the leads, and on the effects of nonequilibrium vibrons. The results are compared with DFT+NGF approach and available experiments.

TT 35.6 Thu 15:15 H21

Transport properties of CuPc based devices — ●COSIMA SCHUSTER — Institut für Physik, Universität Augsburg, D-86135 Augsburg

Electronic structure and transport properties of copper phthalocyanine (CuPc), an organic semiconductor, are investigated using density functional theory and scattering theory combined with the nonequilibrium Green's function formalism. Mainly, we discuss the electronic structure of CuPc attached to gold chains – a prototypical one-dimensional molecular device, which shall serve as model system for CuPc in transistors. There, electron and hole transport with different transport properties is observed depending on the geometry and material of the contacts.

We determine, in particular, the charge density of the HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital). Attached to leads, electrons are transferred to/from the leads, respectively, depending on the contact geometry. For a planar contact, the electron transport properties are intrinsic properties of the molecule and do not depend on the contact material. On the other hand, molecules contacted in perpendicular geometry, become hole doped. The transmission through the molecules does not only depend on the density of states of the device but also on the spatial overlap of molecular orbitals and leads in transport direction. Molecular orbitals localized on the Cu thus do not contribute to the transport in planar geometry, but provide the transmitting channel in perpendicular contact geometry.

TT 35.7 Thu 15:30 H21

Dynamical bi-stability in single-molecule junctions: the case of PTCDA on Ag(111) — ●THOMAS BRUMME¹, OLGA NEUCHEVA², CORMAC TOHER¹, RAFAEL GUTIÉRREZ¹, STEFAN TAUTZ², and GIANAURELIO CUNIBERTI¹ — ¹Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, 01062 Dresden, Germany — ²Institute for Bio- and Nanosystems-3 and JARA-Fundamental of Future Information Technology, Forschungszentrum Juelich GmbH, 52425 Juelich, Germany

A broadly observed phenomenon in experiments on molecular junctions is time dependent switching of the tunneling current. In many cases such behavior involves different current states which are attributed to the transfer of single atoms or functional groups in a molecule between different stable configurations. We describe here the investigation of the current switching observed in a molecular junction formed by a PTCDA molecule between an STM tip and an Ag(111) surface, which is believed to be due to the carboxylic oxygen atom switching between the surface and the tip. The switching process displays a strong dependence on the applied bias voltage between tip and surface, but a much weaker sensitivity to the bias polarity. We analyse the experimental data in terms of a minimal model Hamiltonian approach describing the coupling between an adsorbate level (a relevant PTCDA molecular orbital) to local vibrations excited by the tunneling electron. The switching rates as a function of the applied bias could be fitted with a physically reasonable parameter set.

15 min. break

TT 35.8 Thu 16:00 H21

Spin degrees of freedom in single-molecule junctions — ●STEFAN WAGNER¹, STEFAN BALLMANN¹, DANIEL SECKER¹, FRANK SCHRAMM², MARIO RUBEN², and HEIKO B. WEBER¹ — ¹Lehrstuhl für Angewandte Physik, Universität Erlangen-Nürnberg, Staudtstr. 7/A3, D-91058 Erlangen, Germany — ²Institut für Nanotechnologie, Forschungszentrum Karlsruhe, D-76021 Karlsruhe, Germany

We report on transport measurements on single molecules with spin degrees of freedom.

Fe²⁺-bis(pyrazolyl)pyridine is known as a spin transition compound, in which the Fe²⁺ atom carries either no spin or $S=2$, depending on its local environment. We find qualitatively different zero bias anomalies at very low temperatures, in particular a Kondo feature, but also an unconventional dip-like feature in dI/dV . The anomalies and their behavior in magnetic fields are explained by singlet-triplet transitions and magnetic anisotropy.

In addition, a Bis(Co²⁺)bipyrimidine-wire is investigated, with two Co²⁺ ions. Here, we see two states in the low-bias anomaly, referring to a singlet and multiplet states of the ion pair.

TT 35.9 Thu 16:15 H21

Single-molecule Junctions with C₆₀ Fullerene-based Anchoring Groups — ●CHRISTIAN SEILER¹, ALEXEI BAGRETS², VELIMIR MEDED², and FERDINAND EVERS^{1,2} — ¹Institut für Theorie der Kondensierten Materie, Karlsruhe Institute of Technology (KIT), 76128 Karlsruhe, Germany — ²Institut für Nanotechnologie, Karlsruhe Institute of Technology (KIT), 76021 Karlsruhe, Germany

We investigate electronic structure and recently measured [1] ballistic transport properties of the 1,4-bis(fullero[c]pyrrolidin-1-yl)benzene (BDC60) molecule with gold contacts using Density Functional Theory and the Landauer-Büttiker formalism. We study the potential of C₆₀ fullerenes as anchoring groups from a theoretical point of view. Because the high symmetry of C₆₀ leads to a large HOMO-LUMO gap, it behaves like a band insulator, casting doubts on its suitability as an anchoring group. We analyze the effect of symmetry breaking in the presence of electrodes and discover that the electronic level splitting of C₆₀ is small, not mitigating the issue. Moreover, electronic structure calculations of BDC60 reveal that its current-carrying HOMO is a state originating from the internal benzene ring with an energy in the gap of the anchoring C₆₀. Its hybridization is low which we attribute to the π -systems of C₆₀ and the internal benzene ring being nearly orthogonal. We conclude that this creates an additional tunneling barrier which is in agreement with the low conductance values found in [1].

[1] Christian A. Martin et al., JACS, 130(40):13198-13199, 2008

TT 35.10 Thu 16:30 H21

Atomistic control and stability of molecular junctions: *ab initio* structure prediction via transport observables — ●FLORIAN PUMP, CORMAC TOHER, and GIANAURELIO CUNIBERTI — Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, 01062 Dresden, Germany

The electronic transport properties of molecular junctions are very sensitive to the molecular structure and the contact configuration. We present, using the nonequilibrium Green function formalism in combination with DFT [1], the results of our calculations related to recent experiments investigating the interplay of contact structure and elec-

tronic transport characteristics of molecular systems. We focus on STM-experiments involving single PTCDA [2] and fullerene molecules and on investigations of the long-term behavior of the transport properties of terphenylene and terthiophene molecules in mechanically controllable break-junctions [3]. The STM-setup allows for an enhanced control over the contact geometry using the tip as one of the electrodes which enables the investigation of both the electronic and geometric structure of the junction. Our calculations allow for the systematic investigation of the effect of the contact geometry and the molecular configuration on the transport properties of molecular junctions, which should in turn improve the agreement between theory and experiment.

[1] A. Pecchia and A. Di Carlo, Rep. Prog. Phys. **67**, 1497 (2004).

[2] F. Pump *et al.*, Appl. Phys. A **93**, 335 (2008).

[3] D. Dulic, F. Pump, S. Campidelli, P. Lavie, G. Cuniberti, and A. Filoramo, Ang. Chem. Int. Ed. **48**, 8273 (2009).

TT 35.11 Thu 16:45 H21

Influence of substituents on the transport properties of molecular junctions — ●FALCO HÜSER¹, FABIAN PAULY¹, and GERD SCHÖN^{1,2} — ¹Institut für Theoretische Festkörperphysik, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany — ²Institut für Nanotechnologie, Karlsruhe Institute of Technology (KIT), 76344 Eggenstein-Leopoldshafen, Germany

We study by DFT-based methods the correlation between the Hammett constant and the transport properties of single-molecule junctions, i.e. their conductance and thermopower [1]. Motivated by recent experiments [2,3,4], we focus on a large variety of benzenediamines [2] and of benzene-alkane-dithiols [3].

Electron-donating side-groups shift the conducting molecular orbitals up and increase the conductance and thermopower for the hole-conducting molecules, while electron-accepting side-groups have the opposite effect. By means of a single-level model, we relate these effects to properties of the isolated molecules. Relative variations are found to be in good agreement with the measurements.

Going beyond the experiments, we show how the influence of the substituents can be diminished by alkyl spacers and reestablished by alkenyl spacers.

[1] F. Hüser, F. Pauly, G. Schön (submitted).

[2] L. Venkataraman *et al.*, Nano Lett. **7**, 502 (2007).

[3] E. Leary *et al.*, Chem. Commun. **38**, 3939 (2007).

[4] K. Baheti *et al.*, Nano Lett. **8**, 715 (2008).

TT 35.12 Thu 17:00 H21

Investigating Electron Correlation Effects in Transport in Molecular Junctions — ●CORMAC TOHER, FLORIAN PUMP, and GIANAURELIO CUNIBERTI — Institute for Materials Science, TU Dresden, D-01062 Dresden, Germany.

The use of theoretical and computational methods to understand molecular electronics experiments has been hindered by the fact that the geometric structure of the molecular junction is usually not very well understood. This problem can be overcome by using an STM tip to form one of the electrodes of the junction [1,2], and hence taking advantage of the STM's imaging capabilities to characterize the junction. Here we present the results of recent calculations related to a set of STM experiments studying PTCDA on metallic surfaces, using the non-equilibrium Green function formalism in combination with DFT [3, 4]. This comparison demonstrates that DFT based transport calculations are insufficient to accurately describe such systems, even when the geometric configuration is well-controlled. This is due to the failure of approximate exchange-correlations to describe phenomena such as the derivative discontinuity and the Kondo effect.

[1] F. Pump, R. Temirov, O. Neucheva, S. Soubatch, S. Tautz, M. Rohlfing, and G. Cuniberti, Appl. Phys. A **93**, 335 (2008).

[2] R. Temirov, A. Lassise, F.B. Anders, and F.S. Tautz, Nanotechnology **19**, 065401 (2008).

[3] J.M. Oeler *et al.*, J. Phys. Cond. Matter **14**, 2745 (2002).

[4] A.R. Rocha *et al.*, Phys. Rev. B **73**, 085414 (2006).

TT 35.13 Thu 17:15 H21

Controlling AC transport in graphene-based Fabry-Perot devices using magnetic fields — ●CLAUDIA GOMES DA ROCHA¹, LUIS E. F. FOA TORRES², ANDREA LATGE³, and GIANAURELIO CUNIBERTI¹ — ¹Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, D-01062 Dresden, Germany — ²FaMAF, Universidad Nacional de Cordoba, Ciudad Universitaria 5000 Cordoba, Argentina — ³Instituto de Física, Universidade Federal Fluminense, 24210-340 Niterói-RJ, Brazil

We report on a theoretical study of the effects of time-dependent fields on electronic transport through graphene nanoribbon devices. The Fabry-Perot interference pattern is modified by an ac gating in a way that depends strongly on the shape of the graphene edges. While for armchair edges the patterns are found to be regular and can be controlled very efficiently by tuning the ac field, samples with zigzag edges exhibit a much more complex interference pattern due to their peculiar electronic structure. These studies highlight the main role played by geometric details of graphene nanoribbons within the coherent transport regime. We also extend our analysis to noise power response, identifying under which conditions it is possible to minimize the current fluctuations as well as exploring scaling properties of noise with length and width of the systems. Finally, the response of the system under the presence of an external magnetic field is also analysed in which both DC and AC Fabry-Perot patterns and energy gaps can be efficiently tuned by such controlling parameter.

TT 35.14 Thu 17:30 H21

The dark side of benzene: interference vs. interaction — DAN BOHR¹ and PETER SCHMITTECKERT² — ¹Department of Physics, University of Basel, Switzerland — ²Institut für Nanotechnologie, Karlsruher Institut für Technologie (KIT), Germany

We present the study of the linear conductance vs. applied gate voltage for an interacting six site ring structure, which is threaded by a flux of π and coupled to a left and a right lead. This ring structure is designed to have a vanishing conductance for all gate voltages and temperatures provided interactions are ignored. Therefore this system is an ideal testbed to study the interplay of interaction and interference. First we find a Kondo type resonance for rather large hopping parameter. Second, we find additional resonance peaks which can be explained by a population blocking mechanism. To this end we have to extend the Kubo approach within the Density Matrix Renormalization Group method to handle degenerate states.