

## TT 23: Transport: Nanoelectronics III - Molecular Electronics

Time: Wednesday 9:30–12:30

Location: HSZ 105

TT 23.1 Wed 9:30 HSZ 105

**Length-dependent conductance and thermopower in single-molecule junctions of dithiolated oligophenylene derivatives: A density functional study** — ●FABIAN PAULY<sup>1,2</sup>, JANNE VILJAS<sup>1,2</sup>, and JUAN CARLOS CUEVAS<sup>3</sup> — <sup>1</sup>Institut für Theoretische Festkörperphysik, Universität Karlsruhe, Germany — <sup>2</sup>Forschungszentrum Karlsruhe, Institut für Nanotechnologie, Germany — <sup>3</sup>Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, Spain

We study theoretically the length dependence of both conductance and thermopower in metal-molecule-metal junctions made up of dithiolated oligophenylenes contacted to gold electrodes [1]. We find that while the conductance decays exponentially with increasing molecular length, the thermopower increases linearly as suggested by recent experiments [2]. We also analyze how these transport properties can be tuned with methyl side groups. Our results can be explained by considering the level shifts due to their electron-donating character as well as the tilt-angle dependence of conductance and thermopower. Qualitative features of the substituent effects in our density functional calculations are explained using a tight-binding model. In addition, we observe symmetry-related even-odd transmission channel degeneracies as a function of molecular length.

[1] F. Pauly, J. K. Viljas, and J. C. Cuevas, Phys. Rev. B 78, 035315 (2008).

[2] P. Reddy, S.-Y. Jang, R. A. Segalman, and A. Majumdar, Science 315, 1568 (2007).

TT 23.2 Wed 9:45 HSZ 105

**Opto-electronic properties of gold nanoparticle arrays** — ●CHRISTOPH WEISS, MARKUS A. MANGOLD, and ALEXANDER W. HOLLEITNER — Walter Schottky Institute, Technische Universität München, Am Coulombwall 3, 85748 Garching, Germany

The field of plasmonics has drawn a lot of interest in recent years. It deals with the interaction of metal clusters with light. Special attention was given to the fact that the exciting electric field is strongly enhanced at the surface of metal nanoparticles. Another topic where substantial research efforts are undertaken is the field of molecular electronics. Our research combines the two fields by examining the interaction of light with metal-molecule junctions. To this end, ordered hexagonal arrays of alkanethiol coated gold nanoparticles are produced by self-assembly. In such an array the nanoparticles are separated by nanometer-sized gaps. The nanoparticle arrays are manipulated with a focused ion beam and their opto-electronic properties are probed in the UV and visible spectrum. The strong plasmonic absorption of the nanoparticles can be readily observed in the photoresponse of arrays with alkanethiol coated gold nanoparticles.

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TT 23.3 Wed 10:00 HSZ 105

**Conductance of DNA molecular wires: bridging molecular dynamics and model Hamiltonians** — ●BENJAMIN WOICZIKOWSKI<sup>2</sup>, RAFAEL GUTIERREZ<sup>1</sup>, RODRIGO CAETANO<sup>1</sup>, TOMAS KUBAR<sup>2</sup>, MARCUS ELSTNER<sup>2</sup>, and GIANAURELIO CUNIBERTI<sup>1</sup> — <sup>1</sup>Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, D-01062 Dresden, Germany — <sup>2</sup>Institute for Physical and Theoretical Chemistry, Braunschweig University of Technology, D-38106 Braunschweig, Germany.

We present a hybrid method based on a combination of quantum/classical molecular dynamics (MD) simulations [1] and a model Hamiltonian approach to describe charge transport through biomolecular wires.[2] Our approach maps the molecular electronic structure (obtained from the MD simulations) onto a tight-binding model. The latter is then coupled to a bosonic bath which describes fluctuation effects from the solvent and from the conformational dynamics. We apply this approach to the case of pG-pC and pA-pT oligomers as typical cases. We show that conformational fluctuations are crucial in determining charge transport. Especially, our results indicate that pA-pT shows a much larger current than pG-pC, in contrast to transport calculations performed on static configurations.

[1] T. Kubar, P. B. Woiczikowski, G. Cuniberti, and M. Elstner, J. Phys. Chem. B 112, 7937 (2008).

[2] R. Gutierrez, R. Caetano, B. Woiczikowski, T. Kubar, M. Elstner, and G. Cuniberti, submitted (2008).

TT 23.4 Wed 10:15 HSZ 105

**Screw motion of a DNA duplex during translocation through a nanopore: A coarse-grained model** — ●RAFAEL GUTIERREZ<sup>1</sup>, JEWGENI STARIKOV<sup>2</sup>, DIRK HENNIG<sup>3</sup>, HIROAKI YAMADA<sup>4</sup>, GIANAURELIO CUNIBERTI<sup>1</sup>, and BENGT NORDEN<sup>5</sup> — <sup>1</sup>Institute for Materials Science, Dresden University of Technology, D-01062 Dresden, Germany — <sup>2</sup>Institute for Theoretical Solid State Physics, University of Karlsruhe, D-76131 Karlsruhe, Germany — <sup>3</sup>Institute for Physics, Humboldt University of Berlin, D-12489 Berlin, Germany — <sup>4</sup>Yamada Physics Research Laboratory, Niigata 950-2002, Japan — <sup>5</sup>Department of Physical Chemistry, Chalmers University of Technology, SE-412 96, Gothenburg, Sweden

Based upon the structural properties of DNA and their counterion-water surrounding in solution, we have introduced a screw model describing DNA translocation through artificial nanopores in a qualitatively correct way.[1] This model represents DNA as a "screw", whereas the counterion-hydration shell is a "nut". When an electrical potential is applied across a membrane with a nanopore, the "screw" and "nut" begin to move with respect to each other, so that their mutual rotation is coupled with their mutual translation. As a result, there are peaks of electrical current connected with the mutual translocation of DNA and its counterion-hydration shell, if DNA has some non-regular base-pair sequence. The calculated peaks of current strongly resemble those observed in the pertinent experiments.

[1] E. B. Starikov, D. Hennig, H. Yamada, R. Gutierrez, G. Cuniberti, and B. Norden, submitted (2008)

TT 23.5 Wed 10:30 HSZ 105

**Charge-memory polaron effect in molecular junctions** — ●DMITRY A. RYNDYK<sup>1</sup>, PINO D'AMICO<sup>1</sup>, GIANAURELIO CUNIBERTI<sup>2</sup>, and KLAUS RICHTER<sup>1</sup> — <sup>1</sup>Institute for Theoretical Physics, University of Regensburg, D-93040 Regensburg, Germany — <sup>2</sup>Institute for Material Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, D-01062 Dresden

The charge-memory effect, bistability and switching between charged and neutral states of a molecular junction, as observed in recent STM experiments, is considered within a minimal polaron model. We show that in the case of strong electron-vibron interaction the rate of spontaneous quantum switching between charged and neutral states is exponentially suppressed at zero bias voltage but can be tuned through a wide range of finite switching timescales upon changing the bias. We further find that, while junctions with symmetric voltage drop give rise to random switching at finite bias, asymmetric junctions exhibit hysteretic behavior enabling controlled switching. Lifetimes and charge-voltage curves are calculated by the master equation method for weak coupling to the leads and at stronger coupling by the equation-of-motion method for nonequilibrium Green functions.

TT 23.6 Wed 10:45 HSZ 105

**Spin-Vibration Coupling and Kondo Effect in Transport through Single-Molecule Magnets** — ●FALK MAY<sup>1</sup>, MAARTEN R. WEGEWIJS<sup>2</sup>, and WALTER HOFSTETTER<sup>1</sup> — <sup>1</sup>Institut für Theoretische Physik, J. W. Goethe-Universität, D-60438 Frankfurt, Germany — <sup>2</sup>Institut für Theoretische Physik, Lehrstuhl A, RWTH Aachen, D-52056 Aachen, Germany

We study transport properties of a single-molecule magnet (SMM) strongly coupled to electronic leads in the low temperature limit using the numerical renormalization group. These molecules can be characterized by a large spin and magnetic anisotropies arising from spin-orbit coupling of magnetic ions and ligand field effects. We address the important question of the coupling of the spin to a localized molecular vibration, which can also be anisotropic. We find a rich interplay with effects of intrinsic quantum spin-tunneling, responsible for the Kondo effect in such systems [1], for example a Kondo resonance induced solely by zero-point quantum vibrations.

[1] C. Romeike et al., Phys. Rev. Lett. 96, 19 (2006)

## 15 min. break

TT 23.7 Wed 11:15 HSZ 105

**Vibrational Nonequilibrium Effects in Single-Molecule Conductance** — ●RAINER HÄRTLE and MICHAEL THOSS — Theoretische Chemie, Lichtenbergstrasse 4, D-85747 Garching, Germany

Vibrational nonequilibrium effects in charge transport through single-molecule junctions are theoretically investigated by employing a master equation and a nonequilibrium Green's function approach. For molecules that are asymmetrically bound to the leads, vibrationally coupled charge transport, in particular through a single non-degenerate electronic level, exhibits pronounced rectification and negative differential resistance, which can be solely attributed to vibrational nonequilibrium and electron-hole pair creation processes. Focusing on molecular bridges with multiple electronic states, it is shown that electronic-vibrational coupling triggers a variety of vibronic emission and absorption processes, which influence the conductance properties and mechanical stability of single-molecule junctions profoundly. These processes are analyzed in detail for generic model systems of molecular junctions and for more realistic models that are based on ab-initio quantum chemistry calculations.

TT 23.8 Wed 11:30 HSZ 105

**Entangled excitation dynamics in biomolecules due to spatially correlated environmental fluctuations** — ●PETER NALBACH and MICHAEL THORWART — Freiburg Institute for Advanced Studies, Albert-Ludwigs Universität Freiburg, Albertstr. 19, 79104 Freiburg

In photosynthetic light harvesting complexes, absorbed photons induce excitations of biomolecular chromophores. The excitation moves until it reaches the reaction center where it transfers its energy to chemical reaction energy. A single excitation in two chromophores can be described by a quantum two level system (TLS) where the chromophore coupling is of Förster-type and is described as tunnelling between the two sites. In order to compare the energy transferred to a reaction center with the loss to the environment (formed by vibrational and rotational states of the molecular complexes or charge fluctuations in the solvent-protein host), a coupling of the TLS to an environment is introduced. The environment is characterized by a rather small reorganization energy, leading to slow bath-induced fluctuations. We investigate two coupled TLSs and show that quantum coherent energy transfer is efficiently supported by a slow (non-Markovian) bath which has spatial correlations extending over distances larger than the biomolecule distance. We discuss the inter-chromophore coupling generated by the slow bath modes which couple to both TLSs, its dependence on spatial distance and its influence on the coherent dynamics of the excitations.

TT 23.9 Wed 11:45 HSZ 105

**Phthalocyanine based molecular switches** — ●THOMAS BRUMME, CORMAC TOHER, FLORIAN PUMP, and GIANAURELIO CUNIBERTI — Institute for Materials Science, TU Dresden

Me-Phthalocyanine (MePc) molecules have potential applications as switches in molecular electronics. Recent experiments performed by the group of R. Berndt at the University Kiel indicate that SnPc can be changed between an in-plane and an out-of-plane conformation by means of STM induced tunneling. To control this switching it is crucial to understand the underlying transport mechanism.

We have studied the electronic transport properties of structural conformations of various MePc molecules on a Au(111) surface. In particular, the possibility of switching the larger metal atoms in an STM experiment between an in-plane and an out-of-plane conforma-

tion has been investigated. We used an STM-tip/molecule/surface configuration in order to work out which transport regime could induce this shift. The electronic transport properties and the geometry of the system were calculated for different applied bias voltages. We also investigated the variation of the transport characteristics through the MePc molecule with increasing current.

TT 23.10 Wed 12:00 HSZ 105

**Interplay of Mechanical and Electrical Degrees of Freedom in Molecular Junctions** — ●FLORIAN PUMP<sup>1</sup>, CORMAC TOHER<sup>1</sup>, RUSLAN TEMIROV<sup>2</sup>, OLGA NEUCHEVA<sup>2</sup>, SERGUEI SOUBATCH<sup>2</sup>, STEFAN TAUTZ<sup>2</sup>, MICHAEL ROHLFING<sup>3</sup>, and GIANAURELIO CUNIBERTI<sup>1</sup> — <sup>1</sup>Institute for Materials Science, Dresden University of Technology, D-01062 Dresden — <sup>2</sup>Institut für Bio- und Nanosysteme 3, JARA, Forschungszentrum Jülich, D-52425 Jülich — <sup>3</sup>Department of Physics, University of Osnabrück, D-49069 Osnabrück

The electronic transport properties of molecular junctions are very sensitive to the contact configuration which is usually not very well known. One type of setup which allows more control over the contact geometry uses an STM (scanning tunnelling microscope) tip as one of the electrodes enabling the investigation of both the electronic and geometric structure of the junction. We present, using the nonequilibrium Green function formalism in combination with DFT [2-4], the results of our recent calculations related to a set of experiments studying PTCDA on metallic surfaces [1]. The calculations allow for the systematic investigation of the effect of the contact geometry on the transport properties of molecular junctions, which should in turn improve the agreement between theory and experiment.

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

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

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

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

TT 23.11 Wed 12:15 HSZ 105

**Control of the Conductance in Molecular Switch Junctions** — ●DALIRO NOZAKI, CORMAC TOHER, FLORIAN PUMP, and GIANAURELIO CUNIBERTI — Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, Dresden, Germany

We explore the usability of molecules with bistable characteristics as reversibly tunable molecular switches driven by external stimulation such as light [1] or current-pulse [2]. We have modeled three molecular switch-junctions formed with silicon contacts and azobenzene derivatives which have bistable *cis*- and *trans*-conformations. Using the nonequilibrium Green's function approach implemented with the density-functional-based tight-binding theory [3], we analyzed electron transmission, on/off ratios, potential energy surfaces along reaction coordinate from *cis*- to *trans*-conformation, and the stability of the molecular switches in ambient conditions along MD pathways.

The numerical results have shown that transmission spectra in *cis*-conformations are more conductive than *trans*-ones inside of the bias window in three models. I-V characteristics also lead to the same trends. Additionally, the transmission along MD pathways have shown that the *cis*-conformations are always more conductive than *trans*-ones at room temperature. Therefore, the azobenzene derivative-based molecular switches can be expected to work as robust switching components.

[1] M. del Valle *et al.*, *Nature Nanotech.* **2**, 176 (2007).

[2] H. Riel *et al.*, *Small*, **2**, 973 (2006).

[3] A. Pecchia *et al.*, *Rep. Prog. Phys.* **67**, 1497 (2004).