TT 57: Transport: Molecular Electronics I

Time: Wednesday 9:30–13:00 Location: HSZ 304

TT 57.1 Wed 9:30 HSZ 304

Electrical transport through ferrocene molecules using MCBJ — •Karthiga Kanthasamy, Christoph Tegenkamp, and Herbert Pfnür — Institut für Festkörperphysik (ATMOS), Leibniz Universität Hannover

In order to understand the electrical behavior of molecules we fabricate a stable atomic gold contact by Mechanically Controllable Break Junction(MCBJ). We study the electrical transport through ferrocenedithiol (FDT) molecules because of its rotational flexibility with respect to two cyclopentadienyl(Cp) rings without activation threshold and high conductance.

The molecules are chemisorbed on the Au atomic contact, as confirmed by XPS characterization on Au thin film. After insertion of FDT molecules, measuring conductance versus contact distance at constant voltage 1 mV there are characteristic stepwise changes below 1 G_0 with a very pronounced plateau at 0.4 G_0. In the range 0.1 $G_0 <$ conductance $< 0.4\ G_0$, we measured IV-curves at room temperature and 80 K. Detailed analysis of IV-curves exhibit characteristic peak in the first derivative. The voltage at the maxima can be identified with the excitation of C-H bending, C-H stretching vibrational modes indicating resonantly enhanced conductance by vibrational excitation. The influence of different anchoring groups in the electronic transport will be shown.

TT 57.2 Wed 9:45 HSZ 304

Multi-orbital STM theory for π-conjugated molecules on thin insulating films — •Benjamin Siegert, Andrea Donarini, and Milena Grifoni — Institut für Theoretische Physik, Universität Regensburg, D-93040 Regensburg

We present a multi-orbital extension of our STM theory [1,2,3], based on the reduced density matrix formalism, which is capable of describing electronic transport through π-conjugated molecules on thin insulating films. In order to investigate spin-dependent transport effects using the example of a Cu-Phthalocyanine molecule, we set up an effective manybody Hamiltonian including exchange and spin-orbit interaction. The transport dynamics is calculated by evaluating a Generalized Master Equation including all diagonal and off-diagonal elements of the reduced density matrix of the molecule. Examples are shown of how many-body effects, like interference between degenerate many-body states, can affect the transport properties of π-conjugated molecules on thin insulating films in an STM setup.

[1] S. Sobczyk, A. Donarini, and M. Grifoni, PRB **85**, 205408 (2012).
[2] A. Donarini, B. Siegert, S. Sobczyk, and M. Grifoni, PRB **86**, 155451 (2012).
[3] B. Siegert, A. Donarini and M. Grifoni, pss (b) **250**, 2444 (2013)

TT 57.3 Wed 10:00 HSZ 304

Gate-controlled shift of CoPc orbitals on graphene in a STM junction — •Samuel Bouvron[1], Philipp Erler[1], Alexander Graf[1], Romain Maurand[2], Luca Gragnaniello[1], Dirk Wiedmann[1], Fabian Pauly[1], and Mikhail Fonin[1] — [1]Fachbereich Physik, Universität Konstanz — [2]Department of Physics, University of Basel

One of the most promising aims of molecular electronics is the fabrication of a molecular transistor. Such a device necessarily requires a gate-electrode, which allows to set the molecule into a controllable electric field, therefore tuning the energy of the molecular orbitals relevant for charge transport. Moreover since the very first year of the development of the scanning tunneling microscope (STM), it was proposed to use its extremely high lateral resolution to study molecules and record $I(V)$ or $I(z)$ characteristics of the tunnel contacts formed. However, the geometric constrains of the microscope make the implementation of a third gate-electrode challenging. Here we investigate the electronic properties of cobalt phthalocyanine (CoPc) molecules on graphene on SiO_2/Si, where graphene and Si serve as drain and gate electrode, respectively. We report on site-dependent and gate-dependent tunnel transport through the CoPc molecules. The influence of the gate voltage on the energy position of the molecular orbitals is discussed and the orbital structure is assigned. Finally this STM-tip/molecule/graphene/SiO_2/Si device geometry opens a possibility to combine the spatial resolution of the STM with the additional control of the electric field commonly used in transport measurements.

TT 57.4 Wed 10:15 HSZ 304

Laser pulse induced transient currents in a molecular junction — •Yaroslav Zelinskyy[1,2] and Volkhard May[1] — [1]Institut für Physik, Humboldt Universität zu Berlin — [2]Bogolubov Institute for Theoretical Physics, National Academy of Science of Ukraine

The investigation of transient phenomena in molecular junctions is of increasing interest. As an example, computations are presented on femtosecond laser pulse excitations and subsequent transient current formation. A single molecule sandwiched between two spherical leads is considered[1,2]. Optical excitation of the leads is accounted for by their collective plasmon excitations which may also couple to the molecular excitations. While this coupling may be strong the electron transfer coupling of the molecule to the leads is considered to be small to stay in the regime of sequential charge transmission. All the calculations of transient currents are performed in the framework of a density matrix theory. As a quantity detectable in the experiment the averaged dc–current resulting from a huge sequence of laser pulses is also calculated . The obtained transient currents are confronted with those induced by voltage pulses[3].

[1] L. Wang and V. May, Phys.Chem.Chem.Phys. 13, 8755 (2011)
[2] Y. Zelinskyy and V. May, Nano Lett. 12, 446 (2012)
[3] Y. Zelinskyy and V. May, J. Chem. Phys. (submitted)

TT 57.5 Wed 10:30 HSZ 304

Theory of molecular junction electroluminescence: plasmon-enhancement and emission narrowing due to multiple molecules — •Yuan Zhang and Volkhard May — Institut für Physik, Humboldt Universität zu Berlin, Newtonstraße 15, D-12489, Germany

Photoemission of a molecular junction formed by up to five individual molecules and placed in the proximity of an Au nanosphere is studied theoretically. Charge transmission through the molecules at a finite applied voltage induces an electronic excitation while an energy transfer coupling to the nanosphere is responsible for plasmon excitation and enhancement effects. The consideration of several molecules and larger currents results in an increased electroluminescence intensity compared to earlier studies [1,2].The used density matrix description accounts for the entanglement of different molecular excitations due to their simultaneous coupling to the Au nanosphere. The obtained emission line narrowring with increasing number of molecules is related to the SPASER mechanism.

[1]Y. Zhang, Y. Zelinskyy, and V. May, J. Chem. C **116**, 25962 (2012)
[2]Y. Zhang, Y. Zelinskyy, and V. May, Phys. Rev. B **88**, 155426 (2013)

TT 57.6 Wed 10:45 HSZ 304

Polarization dependent, surface plasmon induced photoconductance in gold nanorod arrays — Sandra Diefenbach[1], •Lisa Kugler[1], Nadine Erhard[1], Johannes Schopka[1], Alfonso Martin[2], Christoph Karnetzky[1], Daniela Iacopino[2], and Alexander Holleitner[1] — [1]Walter Schottky Institut and Physik-Department, Technische Universität München — [2]Tyndall National Institute, University College Cork, Lee Maltings Complex, Dyke Parade, Cork, Ireland

We report on the photoconductance properties of two-dimensional arrays of gold nanorods which are formed by a combination of droplet deposition and stamping methods. The photoconductance of the nanorod arrays is strongly enhanced through the excitation of longitudinal surface plasmons. Hereby, the photoconductance is polarization dependent with a maximum signal for the electric field of the exciting photon being aligned with the longitudinal axis of the nanorods [1]. We interpret the observations by a plasmonically induced, bolometric enhancement of the conductance, but we also discuss a possible hot-electron emission induced by the plasmons.

We gratefully acknowledge support from the European (FP7) 263091 project HYSENS.

[1] S. Diefenbach et al., arXiv:1311.1002 (2013)

15 min. break.

Invited Talk TT 57.7 Wed 11:15 HSZ 304
Quantum Transport at Molecular Scales — •Ferdinand Evers

— Karlsruhe Institute of Technology, Germany

The quantum transport on molecular scales is a wide and very complex research area. It connects to phenomena in experimental disciplines as diverse as the surface sciences, organic electronics, spintronics, catalysis, electrochemistry, strongly correlated systems and even the life sciences. An important motivation in the sub-discipline Molecular Electronics is to single out a single molecule and use it to investigate in detail aspects of the elementary charge transfer processes involved.

The first part of the presentation offers an overview about recent experimental and theoretical developments and new directions in Molecular Electronics. The second part will embark on effects of molecular magnetism. It will deal with the Kondo-effect in molecular adsorbates and explains, how a non-magnetic molecule (hydrogen-phtalocyanine) brings about a giant magnetoresistance.

TT 57.8 Wed 11:45 HSZ 304

Kondo-effect in binuclear metal-organic molecules — •DIMITRA XENIOTI[1,2], ALEXEJ BAGRETS[1], RICHARD KORYTÁR[1], LEI ZHANG[1,3], FRANK SCHRAMM[1], MÉBAREK ALOUANI[2], MARIO RUBEN[1,2], WULF WULFHEKEL[1,3], and FERDINAND EVERS[1,4] — [1]Institute of Nanotechnology (INT), KIT, Karlsruhe, Germany — [2]Institut de Physique et Chimie des Matériaux de Strasbourg (IPCMS), Strasbourg, France — [3]Physikalisches Institut (PI), KIT, Karlsruhe, Germany — [4]Institut für Theorie der Kondensierten Materie (TKM), Karlsruhe, Germany

Low-temperature STM measurements performed on a binuclear metal-organic molecule (Ni(hfacac)$_2$)$_2$(bpmd) ("Ni$_2$") deposited on a Cu surface reveal that the system undergoes a Kondo effect with $T_K \sim 10$K and a spin located nearby Ni atoms. The physics in play is intriguing, because the molecule does not have anchoring groups, which could be responsible for the formation of a chemical bond with the Cu surface. By comparing experimental data with simulated STM images, we have identified possible adsorption geometries. Our simulations show that some observed STM images and large T_K could be attributed to a distorted "Ni$_2$" complex, which is bound to Cu(001) via the bipyrimidine (bpmd) unit. Other images may be interpreted as arising from molecular fragmentation, suggesting a Ni(hfacac)$_2$ moiety to be seen in the experiment. In the latter case our analysis of the DFT+U (Kohn-Sham) spectral function show that the S=1 type Kondo effect arises mainly due to unpaired electrons populating d_{xy} and d_{z^2} orbitals of Ni^{2+} ion. In the case of "Ni$_2$", our calculations suggest that the Kondo effect originates from a pair of weakly coupled S=1 Ni^{2+} spins.

TT 57.9 Wed 12:00 HSZ 304

Heat dissipation and thermopower in atomic-scale junctions — •FABIAN PAULY — Department of Physics, University of Konstanz, Germany

In this talk, I will present recent combined experimental and theoretical efforts to understand the heat dissipation in atomic-scale junctions [1,2]. Using custom-fabricated scanning probes with integrated nanoscale thermocouples, we find that if the junctions have transmission characteristics that are strongly energy dependent, this heat dissipation is asymmetric (that is, unequal between the electrodes) and also dependent on both the bias polarity and the identity of the majority charge carriers. In contrast, junctions consisting of only a few gold atoms, whose transmission characteristics show weak energy dependence, do not exhibit appreciable asymmetry. Our results unambiguously relate the electronic transmission characteristics of atomic-scale junctions to their heat dissipation properties, establishing a framework for understanding heat dissipation in a range of mesoscopic systems where transport is elastic.

[1] W. Lee, K. Kim, W. Jeong, L. A. Zotti, F. Pauly, J. C. Cuevas, and P. Reddy, Nature 498, 209 (2013).
[2] L. A. Zotti, M. Bürkle, F. Pauly, W. Lee, K. Kim, W. Jeong, Y. Asai, P. Reddy, and J. C. Cuevas, New J. Phys. (accepted); arXiv:1307.8336.

TT 57.10 Wed 12:15 HSZ 304

Structure and conductance analysis of atomic-sized contacts — •MANUEL MATT[1], FABIAN PAULY[1], JUAN CARLOS CUEVAS[2], and PETER NIELABA[1] — [1]University of Konstanz, Department of Physics, 78457 Konstanz, Germany — [2]Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, E-28049 Madrid, Spain

We study the conductance histograms of different metals such as Au and Al. Our theoretical approach combines molecular dynamics simulations of the stretching of atomic-sized wires with the non-equilibrium Green's function formalism based on the tight-binding modelling of the electronic system. As compared to pervious work [1], we consider substantially larger wires and explore different lattice orientations. In combination with experiments we recently provided evidence that one can build a fatigue-resistant two-terminal switch with the reversible rearrangement of single atoms[2].

[1] M. Dreher, F. Pauly, J. Heurich, J. C. Cuevas, E. Scheer, and P. Nielaba, Phys. Rev. B 72, 075435 (2005)
[2] C. Schirm, M. Matt, F. Pauly, J. C. Cuevas, P. Nielaba and E. Scheer, Nature Nanotechnology 8, 645 (2013)

TT 57.11 Wed 12:30 HSZ 304

Large Fluctuation of Conductance in Oligo(phenylene ethynylene)-Based Molecular Junctions — •DAIJIRO NOZAKI[1], CORMAC TOHER[2], and GIANAURELIO CUNIBERTI[1,3,4] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany — [2]Department of Mechanical Engineering and Materials Science, Duke University — [3]Center for Advancing Electronics Dresden (cfAED), TU Dresden, 01062 Dresden, Germany — [4]Dresden Center for Computational Materials Science (DCCMS), TU Dresden, 01062 Dresden, Germany

Oligo(phenylene ethynylene)s (OPEs) are widely used as conductive building blocks in molecular electronics. It is known that a phenylene unit in OPEs has low energy barrier (100 meV) for its rotation along the axis of the molecule. We have modeled an OPE-based molecular junction where an OPE oligomer is connected between conducting leads, and calculated the conductance of the junction using gDFTB code [1]. We show that the conductance at the Fermi energy significantly changes as a function of rotation angle of the phenylene unit yielding the on/off ratio over 10^4. We have also performed the ab initio MD at room temperature and obtained the similar result. This result implies that a special attention needs to be paid to the relationship between conductance and fluctuation of molecular framework.

[1] D. Nozaki, C. Toher, and G. Cuniberti, J. Phys. Chem. Lett. **4**, 4192 (2013)

TT 57.12 Wed 12:45 HSZ 304

A Simple Graphical Model to Predict and to Control of Quantum Interference in T-shaped Molecular Junctions — •DAIJIRO NOZAKI[1], HALDUN SEVINCLI[2], STANISLAV M. AVDOSHENKO[3], RAFAEL GUTIERREZ[1], and GIANAURELIO CUNIBERTI[1,4,5] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany. — [2]Department of Micro- and Nanotechnology, TU Denmark, DK-2800 Kgs. Lyngby, Denmark — [3]School of Materials Engineering, Purdue University, Indiana 47907, USA — [4]Center for Advancing Electronics Dresden (cfAED), TU Dresden, 01062 Dresden, Germany. — [5]Dresden Center for Computational Materials Science (DCCMS), TU Dresden, 01062 Dresden, Germany

Quantum interference (QI) effect in molecular devices introduces an abrupt dip in the transmission spectra, thus it can be used to a series of applications such as molecular switches and molecular thermoelectric devices. For the development of the QI-based molecular devices, it is important to give the guideline to predict and to control the shape and positions of QI in transmission spectra. For this purpose, we present a simple graphical approach (called as "parabolic model") to predict the appearance, shape, and energy position of the QI in transmission spectra for T-shaped molecular devices. We show that this "parabolic model" enables one to visualize the relationship between key electronic parameters and the shape of transmission functions without calculating transmission functions.

[1] D. Nozaki, *et al.*, Phys. Chem. Chem. Phys. **15**, 13951 (2013).