CPP 80: Transport: Molecular Electronics (joint session with TT, CPP, HL, MA, O)

Time: Friday 9:30–12:15 Location: H 0110

CPP 80.1 Fri 9:30 H 0110

Phononic and thermoelectric properties of π-stacked molecular junctions — •Thomas Hellmuth[1,3], Marius Bürkle[2], and Fabian Pauly[1] — [1]Theorie der Nanostrukturen, Universität Konstanz, 78457 Konstanz, Germany — [2]Nanosystem Research Institute, National Institute of Advanced Industrial Science and Technology, Japan — [3]Institut für Theoretische Festkörperphysik, Karlsruhe Institute of Technology, 76131 Karlsruhe,Germany

We present our newly developed approach to compute phonon thermal transport through nanosystems from first principles using density functional theory combined with non-equilibrium Green's function techniques. Combining both electron and phonon transport, we analyze the heat transport and thermoelectric properties of π-stacked paracyclophane molecules contacted to gold electrodes [1]. We show that, depending on temperature, the phononic contribution to the heat conductance is a factor of about 5 larger than the electronic one. By calculating the thermoelectric coefficients in linear response, we estimate the figure of merit ZT of the single-molecule junctions for different molecular lengths and substituents.

[1] M. Bürkle, T. J. Hellmuth, F. Pauly, Y. Asai, submitted.

CPP 80.2 Fri 9:45 H 0110

Conductance and thermopower of C_{82} and endohedral metallofullerene molecular junctions with Au electrodes — •Marius Buerkle[1], See Kei Lee[2], Ryo Yamada[2], Hirokazu Tada[2], and Yoshihiro Asai[1] — [1]AIST, NRI, Tsukuba, Japan — [2]Graduate School of Engineering Science, Osaka University, Japan

By combining STM based conductance and thermopower measurements with first-principle transport calculations we investigate the thermoelectric properties of single C_{82} molecules, and its endohedral metallofullerene (EMF) derivatives $Gd@C_{82}$ and $Ce@C_{82}$ bridging Au electrodes. All three molecular junctions show a comparable conductance of around 0.2 G0 and a negative thermopower indicating electron-like transport through the lowest unoccupied molecular orbital (LUMO). However, for the EMF junctions a much larger thermopower is observed which we can relate to changes in the electronic structure induced by the lanthanide atoms.

CPP 80.3 Fri 10:00 H 0110

A scaling relation in the vibronic contribution to the current noise — •Yoshihiro Asai — AIST, Tsukuba, Japan

The electron-phonon coupling effect on the electric current noise is studied based on the fully self-consistent theory of electron and phonon currents (SCEPC) given in terms of the Keldysh Greens function method [1], which has been successful in describing the local heating phenomena [2] and the temperature dependence of the electric conductance [3,4]. Based on the theoretical result on the noise accompanying the vibronic current, we will discuss a scaling relation between the two quantities derived from the current noise and the electric conductance at finite bias voltage. We found that the scaling relation holds when the dynamics of the electron satisfies a specific condition. We will describe these in the talk.

[1] Y. Asai, Phys. Rev. B 78, 045434-1-24 (2008).
[2] Y. Asai, Phys. Rev. B, 84, 085436-1-7 (2011).
[3] S.-K. Lee, R. Yamada, S.Tanaka, G.-S. Chang, Y. Asai, and H. Tada, ACS Nano, 6, 5078-5082 (2012).
[4] Y. Asai, Phys. Rev. B 86, 201405(R)-1-4 (2012).

CPP 80.4 Fri 10:15 H 0110

Thermo-voltage of nano-thermocouples — •Ayelet Ofarim, Bastian Kopp, Johannes Boneberg, Paul Leiderer, and Elke Scheer — University of Konstanz, Department of Physics, Konstanz, Germany

As the down-scaling of electronic components continues, engineering the devices has become a challenge, in particular in view of energy and heat management. Study of thermoelectric effects in nanostructures gives important additional information about charge transport, also regarding possible life-time limiting phenomena and applications for the conversion of light energy via heat into electrical energy. The scope of this presentation is to gain deep insight into the charge transport mechanism, by studying thermo-voltage effects of metallic atomic-sized contacts [1]. We present the concept for determination of the thermo-voltage of nano-thermocouples, using a novel mechanically-controlled break junction (MCBJ) mechanism. A technique to create and detect a temperature gradient, using laser irradiation, is also presented.

[1] B. Kopp, Z. Yi, D. Benner, F. Q. Xie, C. Obermair, T. Schimmel, J. Bonenberg, P. Leiderer and E. Scheer, Beilstein J. Nanotechnol. 3, 703 (2012).

CPP 80.5 Fri 10:30 H 0110

Quantum interference in thermoelectric molecular junctions: A toy model perspective — •Daijiro Nozaki[1], Stas. M. Avdoshenko[2], Haldun Sevinçli[3], and Gianaurelio Cuniberti[1,4,5] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, Germany — [2]Department of Chemistry and Institute for Computational Engineering and Sciences, University of Texas at Austin, USA — [3]Department of Materials Science and Engineering, Izmir Institute of Technology, Turkey — [4]Dresden Center for Computational Materials Science (DCCMS), TU Dresden, Germany — [5]Center for Advancing Electronics Dresden (cfAED), TU Dresden, Germany

In order to reveal the relationship between the line shape of the transmission spectra affected by quantum interference and the electronic structures, we consider a homogeneous toy model where all on-site energies are identical and model four types of molecular junctions due to their topological connectivities. We systematically analyze their transmission spectra, density of states, and thermoelectric properties. Even without the degree of freedom for on-site energies an asymmetric Fano peak could be realized in the homogeneous systems with the cyclic configuration. We also calculate the thermoelectric properties of the model systems with and without fluctuation of on-site energies. Even under the fluctuation of the on-site energies, the finite thermoelectrics are preserved for the Fano resonance, thus cyclic configuration is promising for thermoelectric applications.

[1] D. Nozaki, H. Sevinçli, S. M. Avdoshenko, G. Cuniberti, J. Appl. Phys. 117, 074308 (2014).

CPP 80.6 Fri 10:45 H 0110

Effect of nonadiabatic electronic-vibrational interactions on the transport properties of single-molecule junctions — •André Erpenbeck[1], Rainer Härtle[2], and Michael Thoss[1] — [1]Institut für Theoretische Physik und Interdisziplinäres Zentrum für Molekulare Materialien (ICMM), Friedrich-Alexander-Universität Erlangen-Nürnberg, Staudtstr. 7/B2, D-91058 Erlangen, Germany — [2]nstitut für theoretische Physik, Georg-August-Universität Göttingen, Friedrich-Hund-Platz 1, D-37077 Göttingen, Germany

The interaction between electronic and vibrational degrees of freedom in single-molecule junctions may result from the dependence of the electronic energies or the electronic states of the molecular bridge on the nuclear displacement. The latter mechanism leads to a direct coupling between different electronic states and is referred to as nonadiabatic electronic-vibrational coupling. Employing nonequilibrium Green's functions in combination with the self-consistent Born approximation, we study the influence of nonadiabatic electronic-vibrational coupling in model molecular junctions. Thereby we distinguish between systems with well separated and quasi-degenerate electronic levels. Our results show that the nonadiabatic electronic-vibrational interaction can have a significant influence on the transport properties [1]. The underlying mechanisms are analyzed with respect to the different signatures of nonadiabatic and adiabatic electronic-vibrational coupling, the relevant transport channels, negative differential resistance and quantum interference effects.

[1] A. Erpenbeck et. al., arXiv:1411.5844 (2014)

15 min. break.

CPP 80.7 Fri 11:15 H 0110

Significant role of end groups in electrical transport through molecules — •Karthiga Kanthasamy[1], Markus Ring[2], Fabian Pauly[2], Christoph Tegenkamp[1], and Herbert Pfnür[1] — [1]Institut für Festkörperphysik, Leibniz Universität, Hannover, Germany — [2]Fachbereich Physik, Universität Konstanz, Germany

Mechanically controllable break junction (MCBJ) technique is used to investigate the electronic properties of ferrocene and phenyl based

molecules with different end groups. Stepwise changes in conductance are observed below 1Go after insertion of the molecules. The junctions are opened in vacuum and IV curves are measured for various distances between the electrodes. Detailed analysis of IV curves shows characteristic peaks in the first-order derivative for ferrocene dithiol (FDT) molecules, which are absent in ferrocene diamine (FDA) and biphenyl dithiol (BPDT). For FDT, in the range of 0.56Go to 0.09Go, there are two symmetric peaks, whose energy difference increases from 60 meV to 160 meV with increasing contact distance. Above 0.56Go or below 0.01Go, symmetric peaks are absent. The FDT molecules show typically a one order of magnitude higher conductance than FDA and BPDT. The IV graph for FDT is linear, i.e., it has metallic characteristics, while FDA and BPDT are dominated by tunneling. Theoretical calculations for the molecules in different configurations between the gold electrodes are performed based on density functional theory and the non-equilibrium Green's function formalism. Both elastic transport properties and inelastic electron tunneling spectra are studied to explain the experimental observations.

CPP 80.8 Fri 11:30 H 0110

Photoinduced transient current through a molecular junction: Effects of lead excitation — •Yaroslav Zelinskyy[1,2], Yoram Selzer[3], and Volkhard May[1] — [1]Institut für Physik, Humboldt Universität zu Berlin, Newtonstraße 15, D-12489 Berlin, Germany — [2]Bogolubov Institute for Theoretical Physics, National Academy of Science of Ukraine, 14-b Metrologichna str., UA-03683, Kiev, Ukraine — [3]School of Chemistry, Tel Aviv University, Ramat Aviv, 69978 Tel Aviv, Israel

Laser pulse induced transient currents through a molecular junction are studied in the framework of a density matrix theory. By focusing on the sequential transport regime two types of lead excitation are considered. Firstly, effects of collective plasmon excitations of the leads and their resonant coupling to molecular excitations are investigated. If such a resonant coupling cannot be realized a second excitation regime would be of interest. Now, the nonequilibrium dynamics of individual lead electrons affect the transient current formation. While a resonant coupling to lead plasmon excitations induces a remarkable current enhancement nonequilibrium electron distributions in the leads determine the transient current mainly by their thermalization process. The theoretical framework described above is used to analyze time-resolved conductance measurements of molecular junctions based on Ferrocene molecules.

[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, Y. Selzer and V. May, Phys. Rev. B (submitted)).

CPP 80.9 Fri 11:45 H 0110

***Ab-initio* model of extended CNT-metal contact** — Artem Fediai[1,2,3], •Dmitry Ryndyk[1,2,3], and Gianaurelio Cuniberti[1,2,3] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany — [2]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [3]Dresden Center for Computational Materials Science (DCCMS), TU Dresden, 01062 Dresden, Germany

Relevant CNT-metal contacts belong to so-called extended type. Current flows from electrodes into CNT in a distributed manner, and contact resistance depends on the contact length. In such circumstances the standard *ab-initio* based transport techniques to calculate electron transport should be modified.

We have developed a special method which allows calculation of transport in the systems with metal-CNT contacts at *ab-initio* level. It takes into account both internal and external parts of the CNT-metal contact and requires simulation of the one principal and two auxiliary atomistic systems. Results of *ab-initio* calculations are then subjected to special treatment and being used in Green function formalism afterwards.

This method was applied to Al-CNT and Pd-CNT extended contacts. Results agree perfectly with existing experimental data being indeed obtained at a purely *ab-initio* level.

CPP 80.10 Fri 12:00 H 0110

Hierarchical Quantum Master Equation Approach to Vibrationally Coupled Electron Transport in Single-Molecule Junctions — •Christian Schinabeck[1], Rainer Härtle[2], and Michael Thoss[1] — [1]Institut für Theoretische Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg, Staudtstr. 7/B2, D-91058 Erlangen, Germany — [2]Institut für Theoretische Physik, Georg-August-Universität Göttingen, Friedrich-Hund-Platz 1, D-37077 Göttingen, Germany

We investigate vibrationally coupled transport in single-molecule junctions using the hierarchical quantum master equation (HQME) approach [1-3]. This method allows a systematic convergence of the reduced dynamics of open quantum systems beyond the traditional Markovian rate equations. Within the HQME framework, two different approaches are presented and compared, which describe the vibrational degrees of freedom as part of the system or the bath, respectively. The methodology is applied to a model molecular junction consisting of a molecular level coupled to fermionic leads as well as a vibrational mode. For this system, the accurate results of the hierarchical quantum master equation approach are compared with Markovian rate equation as well as fourth-order time-nonlocal master equation calculations in different parameter regimes. The convergence properties of the two HQME approaches are analyzed in detail.

[1] Y. Tanimura *et al.*, J. Phys. Soc. Jpn. 75, 082001 (2006).
[2] F. Jiang *et al.*, Phys. Rev. B 85, 245427 (2012).
[3] R. Härtle *et al.*, Phys. Rev. B 88, 235426 (2013).