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**Electron transport in atomic and molecular
junctions**

Summary of the Ph.D. thesis

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Introduction

The invention of the Scanning Tunnelling Microscope (STM) in 1986 has opened a new research field, which is merged into nanoscience. With the development of different types of scanning probe techniques matter can be visualized and even be manipulated at atomic scale today. The behavior of nature at the nanometer scale is in the scope of interest not only for physics but also for chemistry and biology. The spacial structure of the vortex cores in high temperature superconductors, the chemical activity of clusters or the mechanical properties of individual protein molecules can be studied with these local probe techniques.

The physics of atomic and molecular junctions is a special field of nanoscience. Only a few atoms or a molecule provide the connection between two electrodes in these junctions. The size of these junctions is comparable with the Fermi-wavelength of the electrons, thus a quantum mechanical description is required to study the transport, mechanical and chemical properties.

Atomic-sized metallic junctions can be created in a simple and reliable way, by the elongation of a macroscopic sized metallic wire. As the wire is pulled, it becomes narrower and narrower, and if the elongation is stopped before the complete breakage, a constriction with only a few atoms remains in the smallest cross section. The extensive experimental and theoretical investigations during the last decade have provided a comprehensive understanding of the transport properties of atomic-sized metallic junctions [Agrait]. The conductance is mainly determined by the chemical nature of the atoms included: the open conductance channels are related to the valence orbitals. Besides, several mesoscopic effects (like conductance fluctuations, shot noise) show up as significant corrections in the transport behavior due to the wave nature of the electrons.

Increasing attention has been paid to molecular junctions recently. These

studies are motivated by the aim of constructing electronic devices at the molecular level. A properly designed organic molecule can act as a diode, transistor or logical gate, and such molecules are candidates for being the building blocks of a future electronics at the ultimate small scale. At the actual stage of this field there is still a lot to do to understand and control the transport properties of single molecules. The experimental results are usually not completely reproducible, it is not easy to identify the number of molecules connecting the two electrodes. The theoretical models are also far from being satisfactory.

In order to experimentally study the transport properties of atomic and molecular junctions a sample holder implementing the Mechanically Controllable Break Junction (MCBJ) technique was built in the Low Temperature Physics Laboratory of the Department of Physics, BUTE [Halbritter]. Atomic-sized contacts are created by fixing a notched wire on the top of a flexible beam and breaking the wire by bending the beam. The breakage of the wire at cryogenic circumstances naturally provides clean surfaces and the geometry of the setup ensures a large mechanical stability.

Objectives

During the buildup of the MCBJ technique in our laboratory the development of the computer controls of the break junction setup was my task. This software and the associated electronics enables the measurement and the online analysis of various physical phenomena, like conductance histograms, plateaus' length and conductance fluctuation analysis, phonon spectroscopy etc.

The objective of my Ph.D. work was to investigate the transport properties of molecular test systems. Therefore I have studied the interaction of the simplest molecule, H_2 with atomic-sized metallic junctions. Applying

different statistical methods to analyze the conductance traces I have identified new hydrogen-related atomic configurations and I proposed possible pictures for their microscopic origin.

New scientific results

The main scientific results of my Ph.D. work are summarized in the following thesis points.

1. I have developed computer controlled electronic setups for various types of experiments on atomic and molecular junctions, created by the mechanically controllable break junction technique. These instrumentations include interactive measurement control, including regulation, settings, feedback; fast data acquisition, and online statistical analysis of the data. The setups are suitable for precise calibration of the electrode separation on the 0.1 Å level (even for modified work functions due to adsorbed molecules) [4]; determination of vibration modes of mesoscopic junctions [2]; study of correlations between atomic configurations [1,2]; and the analysis of the number of conductance channels in conductance fluctuation measurements [2,3]. As an example, I have shown that the $G=1 G_0$ configuration in palladium-hydrogen system does not have a single perfectly transmitting channel. Instead, more partially open channels contribute to its conduction [2].
2. I have investigated quantum interference corrections in the fine structure of the conductance plateaus in gold nanocontacts applying a novel statistical approach. The contributions of quantum interference and the strain dependence of the local atomic configuration to the plateaus' slope has been separated. The quantitative analysis has shown that the influence of the quantum interference phenomenon and that of the atomic discreteness of the junction are in the same order

of magnitude. Additionally, I have performed a model calculation to estimate the amplitude of the quantum interference-induced fluctuations in the slope of the plateaus. The results of this calculation show good agreement with the experimental findings. [3]

3. I have shown that the hydrogen gas strongly reacts with atomic-sized palladium junctions. In the presence of hydrogen, the peak corresponding to the monoatomic palladium contact disappears from the conductance histogram, and two new peaks emerge. These peaks at $G \sim 0.5 G_0$ and $\sim 1 G_0$ are attributed to two new hydrogen-related atomic configurations. Based on phonon spectrum measurements I have demonstrated that these configurations are situated between electrodes containing dissolved hydrogen atoms. The dissolution of hydrogen makes a crucial difference compared to the isoelectric platinum nanocontacts, where the hydrogen is found only on the surface. Combining the existing results on Pt-H₂ junctions [Heurich] and the consequences of the dissolution of hydrogen on the band structure of the Pd electrodes [Chan], I have given a possible explanation for the microscopic realization of the two new atomic configurations: hydrogen-bridged and Pd-bridged palladium hydride electrodes. [2]
4. I have studied the influence of adsorbed hydrogen on the conductance of gold nanojunctions by novel statistical methods. Both the correlation techniques applied for the length distribution of the plateaus and the investigation of the contact evolution during elongation and retraction have demonstrated that atomic gold chains are formed in H₂ environment, but the transmission is strongly reduced due to interaction with hydrogen. I have found that the conductance traces frequently show a well-defined periodic behavior with positive slope in the tail region. I have shown, by statistical methods, that these curves represent a new type of chain evolution during the elongation,

and I proposed a microscopic explanation for this behavior: a hydrogen molecule gets incorporated in the gold nanojunction and this hydrogen clamp is strong enough to pull a monoatomic gold chain from the electrodes. [1,5]

List of publications

Publications referred in my thesis points:

- [1] Sz. Csonka, A. Halbritter, G. Mihály,
Pulling goldnanowires with a hydrogen clamp.
accepted in Phys. Rev. B, cond-mat/0502421 (2005).
- [2] Sz. Csonka, A. Halbritter, G. Mihály,
O. I. Shklyarevskii, S. Speller, and H. van Kempen
Conductance of Pd-H nanojunctions.
Phys. Rev. Lett., **93**, 016802 (2004).
- [3] A. Halbritter, Sz. Csonka, G. Mihály,
O. I. Shklyarevskii, S. Speller and H. van Kempen
Quantum interference structures in the conductance plateaus of gold nano-junctions.
Phys. Rev. B Rap. Com., **69**, 121411 (2004).
- [4] Sz. Csonka, A. Halbritter, G. Mihály, E. Jurdik,
O. I. Shklyarevskii, S. Speller, and H. van Kempen
Field and temperature induced effects in the surface modification process.
Journal of Applied Physics, **96**, 6169 (2004).
- [5] Sz. Csonka, A. Halbritter, G. Mihály, E. Jurdik,
O. I. Shklyarevskii, S. Speller, and H. van Kempen,
Fractional conductance in hydrogen-embedded gold nanowires.
Phys. Rev. Lett., **90**, 116803 (2003).

Further publications:

[6] I. K. Yanson, O. I. Shklyarevskii, Sz. Csonka, H. van Kempen, S. Speller, A. I. Yanson, and J. M. van Ruitenbeek, *Atomic size oscillations in conductance histograms for Au nanowires and the influence of work hardening*. accepted in Phys. Rev. Lett., cond-mat/0508194 (2005).

[7] A. Halbritter, Sz. Csonka, G. Mihály, E. Jurdik, O. Yu. Kolesnychenko, O. I. Shklyarevskii, S. Speller, and H. van Kempen, *Transition from tunneling to direct contact in tungsten nanojunctions*. Phys. Rev. B, **68**, 35417 (2003).

[8] A. Halbritter, Sz. Csonka, O. Yu. Kolesnychenko, G. Mihály, O. I. Shklyarevskii, and H. van Kempen, *Connective neck evolution and conductance steps in “hot” point-contacts*. Phys. Rev. B, **65**, 45413 (2002).

[9] P. Fazekas, K. Penc, H. Berger, L. Forró, Sz. Csonka, I. Kézsmárki and G. Mihály *BaVS₃: from spin gap insulator to non Fermi liquid*. Physica B, **312**, 694 (2002).

[10] I. Kézsmárki, Sz. Csonka, H. Berger, L. Forró, P. Fazekas and G. Mihály *Pressure dependence of the spin gap in BaVS₃*. Phys. Rev. B Rap. Com., **63**, 81106 (2001).

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[Agrait] N. Agrait, A. L. Yeyati, and J. M. van Ruitenbeek.

Quantum properties of atomic-sized conductors

Phys. Rep., **377**, 81 (2003).

[Heurich] J. Heurich, J. C. Cuevas, F. Pauly, W. Wenzel, and G. Schon.

Theoretical description of the electrical conduction in atomic and molecular junctions.

Nanotechnology, **14**, R29 (2003).

[Chan] C. T. Chan and S. G. Louie.

Self-consistent pseudopotential calculation of the electric structure of PdH and Pd₄H

Phys. Rev. B, **27**, 3325 (1983).

[Halbritter, 2003] A. Halbritter.

Investigation of atomic-sized conductors with the mechanically controllable break junction technique.

Ph.D thesis, Budapest University of Technology and Economics, Hungary (2003).