O 3: New Methods and Developments I: Scanning Probe Techniques

Time: Monday 10:30–13:15 Location: H5

O 3.1 Mon 10:30 H5

Laser Actuation of Micro Cantilevers via Thermo-Mechanical Symmetry Breaking — •Sven Kraft, Boris Hage, Ingo Barke, and Sylvia Speller — Institute of Physics, University of Rostock, 18051 Rostock

Direct actuation of Atomic Force Microscopy (AFM) cantilevers by a laser beam is particularly useful for liquid environments [1]. Various mechanisms are proposed in the literature how laser irradiation leads to mechanical cantilever excitation [2-4]. We present a systematic and comprehensive study of the spatially and frequency resolved excitation efficiency by mapping the oscillation amplitude and phase as a function of the location of an intensity modulated laser. We observe a rich mode structure with surprisingly efficient excitation at the edges, as well as multiple phase reversals in transversal direction of the cantilever, resulting in a checkerboard-like phase pattern. A comparison of coated and uncoated cantilevers, and of illumination from the top and bottom side reveals anisotropic thermal deformation being the dominant mechanism of actuation. This is corroborated by analytical calculations based on solid mechanics which take into account the cross sectional shape of the cantilevers. We also discuss the effect of an internal light intensity structure due to multiple reflection within the cantilever.

O 3.2 Mon 10:45 H5

Lateral manipulation of single iron adatoms by combined atomic force and scanning tunneling microscopy using CO-terminated tips — •Julian Berwanger, Ferdinand Huber, Fabian Stilp, and Franz J. Giessibl — University of Regensburg, 93040 Regensburg, Germany

The functionalization of scanning probe microscope (SPM) tips with a CO molecule is widely used due to their outstanding resolution capability on molecules, metal clusters and other sample systems [1-3]. However, the feasibility of CO tips for an extension of imaging by SPM to controlled atomic manipulation [4] has not been demonstrated. Here, we perform lateral manipulation of single atoms with metal and CO tips. We first study the manipulation characteristics of single atom metal tips and find that asymmetric tips, i.e. tips with a front atom that sits on a tilted atomic tip plane, yield an asymmetry in the lateral force field. When attaching a CO molecule to these tips, their asymmetric force field appears inverted with respect to the underlying tilted metal tips. By applying an analytical model, we propose that the electrostatic interaction is responsible for this inversion of the asymmetry of the lateral force field [5]. Finally we also demonstrate that CO tips can be used to reliably build up large iron clusters atom by atom starting from individual iron atoms.

[1] L. Gross et al. Science 325, 110 (2009); [2] M. Emmrich et al. Science 348, 6232 (2015); [3] M. Ellner et al. Nano Lett. 16, 3 (2016); [4] D. M. Eigler et al., Nature 344, 524 (1990); [5] J. Berwanger et al. PRB 98, 195409 (2018)

O 3.3 Mon 11:00 H5

Constant current scanning mode for imaging single molecules using low temperature atomic force microscopy with CO functionalized tips — •Daniel Martin-Jimenez[1], Doreen Mollenhauer[2], Hermann A. Wegner[3], Andre Schirmeisen[1], and Daniel Ebeling[1] — [1]Institute of Applied Physics (IAP), Justus Liebig University Giessen, Heinrich-Buff-Ring 16, 35392 Giessen, Germany — [2]Institute of Physical Chemistry, Justus Liebig University Giessen, Heinrich-Buff-Ring 17, 35392 Giessen, Germany — [3]Institute of Organic Chemistry, Justus Liebig University Giessen, Heinrich-Buff-Ring 17, 35392 Giessen, Germany

Functionalizing the tip of a low temperature atomic force microscope (AFM) with a single CO molecule facilitates imaging of single adsorbed molecules with submolecular resolution; this is often denoted as bond imaging AFM. Therewith identification of adsorption structures, reaction pathways, etc. becomes feasible. In the standard bond imaging technique, the sensor scans the samples in constant height. While this scanning mode is, in particular, suitable for types of molecules that adsorb planar to the substrate, it is incapable of imaging 3D adsorption structures. To solve this problem, we utilize a scanning mode that achieves submolecular resolution while it tracks the topography by applying a constant current. Constant height and constant current scanning modes are systematically compared and advantages and disadvantages are presented.

O 3.4 Mon 11:15 H5

Probing coherence within tunneling events in a superconducting junction — •Piotr Kot[1], Robert Drost[1], Maximilian Uhl[1], Joachim Ankerhold[2], Alfredo Levy Yeyati[3], Juan Carlos Cuevas[3], and Christian R. Ast[1] — [1]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [2]Institut für Komplexe Quantensysteme and IQST, Ulm, Germany — [3]Departamento de Fisica Teorica de la Materia Condensada, Madrid, Spain

Using a superconducting tunnel junction irradiated by microwaves, we study the coherent and incoherent processes in photon-assisted tunneling. While our junction operates within the dynamical Coulomb blockade regime that involves incoherent tunneling between events, we find coherent processes within a tunneling event that can be tuned by varying the microwave intensity. We find that higher order charge transfers can not necessarily be related to the simple Tien-Gordon equation. Taking the experiment to the limit, we are able to measure coherent tunneling processes involving photons up to 50^{th} order that do not quench superconductivity but rather distribute the spectral weight accordingly. In addition, in the high transmission regime coherence within Andreev reflections is demonstrated. Our finding suggest new avenues of studying higher order charge processes in a single junction.

O 3.5 Mon 11:30 H5

Single switching events of one molecule observed by femtosecond STM — •Dominik Peller, Thomas Buchner, Lukas Kastner, Rupert Huber, and Jascha Repp — University of Regensburg, 93040 Regensburg, Germany

Using THz waves to control electrons in a scanning tunneling microscope (STM) has opened the door to ultrafast atomic-scale microscopy. Recently, we combined low-temperature STM with ultrafast single-electron lightwave electronics to push microscopy to the ultimate spatio-temporal quantum limit via a novel state-selective tunneling regime [1]. The peak of a THz electric-field waveform transiently opens an otherwise forbidden tunneling channel through a single molecular orbital. This process allows us to record the first-ever ∼100 fs sub-Å snapshot images of a single molecule's orbitals and the first femtosecond movie of a single vibrating molecule.

Here, we introduce single-shot detection in lightwave STM as the first approach resolving individual unidirectional quantum events on simultaneous atomic and femtosecond scales. We trigger switching of an individual molecule by single-electron injection and detect every event in real time. Analyzing the quantum statistics of the different reaction paths separately, electron by electron, we time-resolve the molecule's ultrafast, statistical motion along its reaction coordinate.

Moreover, this process allows us to measure a THz near-field waveform within a subatomic volume directly in the time domain.

[1] T. L. Cocker, D. Peller *et al.*, Nature **539**, 263-267 (2016)

O 3.6 Mon 11:45 H5

Nature of binding in planar halogenbenzene assemblies and their possible visualization in scanning probe microscopy — •Aurelio Gallardo[1], Jindrich Fanfrlík[2], Pavel Hobza[2], and Pavel Jelínek[1] — [1]FZU of the CAS, Prague, Czech Republic — [2]IOCB of the CAS, Prague, Czech Republic

High-resolution scanning probe imaging of molecular structures on surfaces with functionalized tips provided the unprecedented spatial resolution. However, the origin of intermolecular features in high-resolution images of molecular assemblies is still under debate. Originally, such features were considered as a direct observation of weak non-covalent bonds between molecules. Nevertheless, this interpretation was challenged and ascribed to an experimental artefact. To address this long-standing controversy, we provided theoretical analysis of intermolecular interaction and high-resolution imaging of halogen substituted benzenes assemblies deposited on metallic substrate, which was extensively studied experimentally. First, we show that formation of molecular assemblies made of C6Br6 and C6F6 on surfaces is driven by interplay between halogen and dispersive interaction. Next, for the C6Br6 and C6F6 assemblies on surface we analyze simulated

high-resolution IETS and AFM images acquired with a CO-tip. Very good agreement with the experimental evidence allows us to unambiguously determine the origin of the sharp edges. We discuss, why such sharp features should not be interpreted as the direct evidence of the signature of weak non-covalent bonds.

Gallardo et al., J. Phys. Chem. C, DOI:10.1021/acs.jpcc.8b09631

O 3.7 Mon 12:00 H5

Controlled Modulation of Plasmonic Response in a Scanning Tunneling Microscope Junction by Fabry Pérot Type Interference — •Hannes Böckmann[1], Shuyi Liu[2], Melanie Müller[2], Adnan Hammud[3], Martin Wolf[2], and Takashi Kumagai[2,4] — [1]University of Göttingen, 37077 Göttingen, Germany — [2]Department of Physical Chemistry, Fritz-Haber Institute, 14195 Berlin, Germany — [3]Department of Inorganic Chemistry, Fritz-Haber Institute, 14195 Berlin, Germany — [4]JST-PRESTO, Kawaguchi, Saitama 332-0012, Japan

We demonstrate that electroluminescence (EL) spectra from a plasmonic scanning tunneling microscope (STM) junction can be manipulated using nanofabricated Au tips by focused ion beam (FIB) milling. A broadband emission spectrum of the localized surface plasmon (LSP) resonance in the STM junction can be periodically modulated by introducing a groove structure on the FIB-polished smooth tip shaft. This groove reflects partially the propagating surface plasmon polariton (SPP) that is generated upon LSP excitation at the tip apex through inelastic electron tunneling, which consequently leads to Fabry Pérot type interference. The SPP also couples again with the LSP mode at the groove and radiates the modulated light. It is found that the emission spectra exhibit a pi phase shift between the apex and the groove, which can be rationalized by the transfer of excitation between the localized LSPs at the apex and groove through the discretized Farby Pérot like SPP levels confined in the tip shaft.

O 3.8 Mon 12:15 H5

Photon-Assisted Tunneling between Superconductors — •Maximilian Uhl, Piotr Kot, Robert Drost, and Christian R. Ast — Max-Planck-Institut für Festkörperforschung, Stuttgart

Microwaves radiated externally into a superconductor tunnel junction of a scanning tunneling microscope influence different kinds of tunneling processes. Here we use a superconducting vanadium tip and sample at a temperature of 300 mK. An external antenna has been designed to produce microwaves in a continuous range from 60 to 90 GHz [2]. It is powered through coaxial cables that are superconducting in the cooled part of the cryostat. The electric field distribution of the antenna radiation depends on the local geometry at the tunnel junction. By emitting and absorbing different numbers of photons, quasiparticles can start tunneling at energies different from the sum of the superconductors' energy gaps [1, 3]. High resolution measurements of photon-assisted tunneling in dependency of bias voltage, microwave voltage and frequency show that there is a strong transmission of the signal to the antenna. The technique of photon-assisted tunneling measurement is applied to quasiparticle as well as Cooper pair tunneling. Furthermore, it opens up new possibilities for electron spin resonance measurements.

[1] G. Falci, V. Bubanja, G. Schön: Z. Phys. B **85**, 451 (1991)
[2] J. Merkt: KIT, Bachelor thesis (2016)
[3] A. Roychowdhury et al.: Phys. Rev. Applied **4**, 034011 (2015)

O 3.9 Mon 12:30 H5

Combining high-resolution Atomic Force Microscopy with Scanning Tunneling Microscopy induced light emission on single molecules — •Katharina Kaiser, Fabian Schulz, and Leo Gross — IBM Research - Zurich, Saeumerstrasse 4, 8803 Rueschlikon

The field of STM induced light emission (STM-LE), especially on single molecules, has grown rapidly in the past 25 years [1, 2] with astounding spatial as well as energetic resolution [3, 4]. Yet, combining structural and optical information on single molecules remains challenging.

We present first results of a combined AFM and STM-LE setup on single vanadyl-phthalocyanine (VOPc) molecules. This setup so far allows for structure determination with atomic resolution by AFM with CO functionalized tips [5] and the possibility to perform controlled atom manipulation conjunct with the investigation of opto-electronic properties by STM-LE.

This work was financially supported by the European Research Council consolidator grant AMSEL.

[1] R. Berndt et al. (1993). Science, 262(5138), 1425-1427.
[2] X. Qiu et al. (2003). Science, 299(5606), 542-547.
[3] B. Doppagne et al. (2017). Phys. Rev. Lett., 118(12), 127401.
[4] A. Yu et al. (2018). Nano Lett., 18(5), 3076-3080.
[5] Gross et al. (2009). Science, 325(5944), 1110-1114.

O 3.10 Mon 12:45 H5

Near-field driven photo-assisted Scanning Tunneling Microscopy — •Benjamin Schröder[1], Ole Bunjes[1], Lara Wimmer[1], Katharina Kaiser[2], Martin Wenderoth[1], and Claus Ropers[1] — [1]Georg-August-Universität, IV. Physikalisches Institut, Friedrich-Hund-Platz 1, 37077 Göttingen — [2]IBM Forschungslabor, Säumerstrasse 4, 8803 Rüschlikon

Recent developments in Scanning Tunneling Microscopy (STM) pave the way towards a controlled optical excitation of the tunnel junction with ultrafast laser pulses. This promises the local observation of surface dynamics, including photochemical reactions, phase transitions and optical manipulations of surface defects on the atomic scale.
Here, we combine an ultra-high-vacuum low-temperature STM with femtosecond laser excitation. The laser is focused directly into the tunneling junction formed by a gold tip and a Cu(100) surface. The strongly enhanced optical near field in the vacuum gap results in an additional photocurrent, evident by comparing current-distance measurements with and without laser illumination. Specifically, current-distance dependencies exhibit a contribution with a spatial decay length about ten times larger than for regular tunneling.
We employ a one-dimensional model to extract an effective energy distribution of tunneling electrons from the experimental data. We discuss contributions from multiphoton excitation and hot electron tunneling.
This project is financially supported by the DFG in the SFB 1073 (project C4).

O 3.11 Mon 13:00 H5

The steep slope to high-resolution MRFM — •Marc-Dominik Krass, Urs Grob, Raphael Pachlatko, Martin Héritier, Jan Rhensius, Alexander Eichler, and Christian Degen — Spin Physics and Imaging, ETH Zurich, Zurich, Switzerland

The goal of nanoscale magnetic resonance imaging (NanoMRI) is the 3D visualization of nuclear spin densities inside objects with near-atomic spatial resolution. One promising candidate for NanoMRI is magnetic resonance force microscopy (MRFM) which employs an ultrasensitive nanomechanical transducer to detect the interaction between nuclear spins and a magnetic field gradient [1]. In recent years, researchers have greatly improved the sensitivity of mechanical transduction [2]. At the same time, other aspects of high-resolution MRFM received little attention, even though they are just as critical.

We identify and analyze fundamental limitations of MRFM resolution and present stringent solutions to them. In particular, we shine light on the role of spin inversion pulses in the presence of thermomechanical motion, cantilever bending, and scanning stage stability. Our work should establish a recipe for subnanometer-resolution MRFM. As a result of our efforts, we demonstrate line scans with a 1D resolution below 2 nm, with a sensitivity corresponding to about 3000 protons.

[1] Christian Degen, et al., Nanoscale magnetic resonance imaging, PNAS 106, 1313 (2009). [2] William Rose, et al., High-Resolution Nanoscale Solid-State Nuclear Magnetic Resonance Spectroscopy, Phys. Rev. X 8, 011030 (2018).