Faster, Smaller, Stronger, Brighter - Advances in Scanning Probe Techniques

756. WE-Heraeus-Seminar

01 Nov - 05 Nov 2021
hybrid at the Physikzentrum Bad Honnef
The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany’s most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the 756. WE-Heraeus-Seminar:

Since their invention in the 1980s, scanning probe microscopy (SPM) methods have become a vital tool for atomic-scale imaging. Remarkably, these techniques, most prominently scanning tunneling microscopy (STM) and atomic force microscopy (AFM), have evolved far beyond the concept of pure imaging devices and nowadays feature a full physico-chemical lab at the nanoscale. The ability to control single atoms or molecules, probe forces between them and study their interactions with electromagnetic fields in both static and time-resolved fashion allows studies of physical processes with unprecedented resolution.

The aim of the WEH seminar ‘Faster, smaller, stronger, brighter - Advances in Scanning Probes Techniques’ is to pedagogically present the recent progress in this rapidly progressing field and provide a platform for extended discussions profitable especially for the younger researchers in the community.

Scientific Organizers:

Dr. Robert Drost 
Aalto University, Finland 
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Introduction

Administrative Organization:

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Venue:

Physikzentrum  
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Registration:

Martina Albert (WE Heraeus Foundation)  
at the Physikzentrum, reception office  
Monday (16:00 h – 20:00 h) and Tuesday morning
Program
# Program

## Monday, 1 November 2021

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<td>from 18:00</td>
<td>BUFFET SUPPER / Informal get together</td>
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<td>20:00</td>
<td>Javier Aizpurua Addressing photonics at the atomic scale</td>
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<td>08:45 – 09:00</td>
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<td>09:00 – 09:45</td>
<td>Susanne Baumann Stochastic resonance as a tool to investigate spin dynamics</td>
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<tr>
<td>09:45 – 10:30</td>
<td>Thomas Fredriksen Spin physics in graphene nanostructures</td>
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<td>10:30 – 11:00</td>
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<td>11:00 – 11:45</td>
<td>Nadine Hauptmann What can atomic force microscopy contribute to understand atomic-scale magnetism?</td>
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<tr>
<td>11:45 – 12:30</td>
<td>Shawulienu Kezilebieke Topological superconductivity in van der Waals heterostructures</td>
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Tuesday, 2 November 2021

14:00 – 15:30  Poster Session A

15:30 – 16:00  COFFEE BREAK

16:00 – 16:45  Tyler Cocker  Atomically resolved terahertz scanning tunnelling spectroscopy as a tool for exploring new materials

16:45 – 17:15  Manish Garg  Real-space sub-femtosecond imaging of quantum electronic coherences in molecules

17:15 – 18:00  Melanie Müller  Tracing ultrafast hot electron dynamics inside a THz-gated STM tip

18:00  DINNER

20:00 – 21:00  Ingmar Swart  Electronic quantum materials simulated with artificial lattices
Program

Wednesday, 3 November 2021

08:00  BREAKFAST

09:00 – 09:45  Guillaume Schull  From single-molecule fluorescence to photosynthesis with an STM

09:45 – 10:30  Yang Zhang  (online)  Real-space evidence for wavelike electronic energy transfer in donor–acceptor molecular systems through quantum coherence

10:30 – 11:00  COFFEE BREAK

11:00 – 11:45  Blanca Biel  (online)  SPM DFT-based simulations of defected 2D materials

11:45 – 12:30  Remy Pawlak  On-surface synthesis of silicene and nanographene characterized by atomic force microscopy

12:30 – 12:40  Conference Photo  (in front of the Physikzentrum/Main entrance)

12:40  LUNCH

14:00 – 18:00  Excursion

18:00  HERAEUS DINNER at the Physikzentrum  
(cold & warm buffet, with complimentary drinks)
## Program

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<td>09:00 – 09:45</td>
<td>Anika Schlenhoff (online)</td>
<td>Using SP-STM at nanometer distances for imaging and manipulation of atomic-scale magnetism</td>
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<td>09:45 – 10:30</td>
<td>Dimas de Oteyza</td>
<td>Chemical stability of zigzag edges in carbon nanostructures</td>
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<td>10:30 – 11:00</td>
<td>COFFEE BREAK</td>
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<td>11:00 – 11:45</td>
<td>Martin Švec</td>
<td>From single-molecule emitters to entangled excitonic states: the insights from the scanning-probe spectromicroscopy</td>
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<td>11:45 – 12:30</td>
<td>Elizabeth Boer-Duchemin</td>
<td>Brighter but not smaller: advances in light emission using scanning probe techniques in air</td>
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<td>12:30</td>
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<td>14:00 – 15:30</td>
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<td>16:00 – 16:45</td>
<td>Katrin Domke</td>
<td>Probing geometric and electronic properties of single-molecule junctions with a combined operando nearfield Raman &amp; break-junction platform</td>
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<td>Philip Willke</td>
<td>Coherent Control of Individual Atomic and Molecular Spins on Surfaces</td>
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<td>17:15 – 18:00</td>
<td>Fernando Delgado</td>
<td>A general description of driven dissipative systems beyond the Bloch equations</td>
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<td>20:00 – 21:00</td>
<td>Fabian Natterer</td>
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<td>08:00</td>
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<td>09:00 – 09:45</td>
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<td>Sifting Self-Organisation: Automated Classification of Far-From-Equilibrium Nanostructures</td>
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<td>09:45 – 10:30</td>
<td>Annica Black-Schaffer (online)</td>
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<td>11:00 – 11:45</td>
<td>Deung-Jang Choi</td>
<td>Creating new bands in a superconductor by crafting different types of spin chains</td>
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<td>Imaging the effect of electron transfer at the atomic scale</td>
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25 Nils Krane  
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26 Felix Küster (online)  
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27 Rian Ligthart  
Artificial atoms on thin Ag layers on Si(111)

28 Benjamin Mallada  
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29 Cristina Mier (online)  
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30 Tomas Neuman  
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36 Rika Simon  
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Abstracts of Lectures

(in alphabetical order)
Addressing photonics at the atomic scale

Javier Aizpurua

Center for Materials Physics in San Sebastian (CSIC-UPV/EHU), Donostia, Spain

A plasmonic nanogap is a superb configuration to explore the interplay between light and matter. Light scattered off, or emitted from a nanogap carries the information of the surrounding electromagnetic environment with it [1,2]. In metallic nanocavities with ultrasmall gaps, electron currents across the gap at optical frequencies efficiently produce a strong nonlinear optical response. All these effects can be further controlled when a bias is applied across the gap, enabling the possibility of active control of light emitted from the cavity [3]. This situation becomes even more appealing when a molecule is located in the gap of the plasmonic cavity or in its proximity [4], with the molecule playing an active role either in the electromagnetic coupling with the cavity, or even participating in processes of charge injection and transfer, which can be revealed through molecular electroluminescence [5]. Here, we will address situations of light emission in electron tunneling configurations where atomic-scale resolution is achieved due to the presence of picocavities within the gap [6]. The process of interaction between a molecular emitter and a tunneling cavity will be addressed both in the weak and strong coupling regimes, as revealed in light absorption and in emission. Strong coupling between a molecule and a plasmonic cavity shows great technological potential as it produces hybrid molecule-cavity polaritonic states which can be used for quantum information or in induced chemical reactivity.

References

Stochastic resonance as a tool to investigate spin dynamics

S. Baumann

University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

Stochastic resonance is an unusual phenomenon in which noise can be used as a resource to synchronize stochastic dynamics to a control signal [1,2]. In this talk, I will show how stochastic resonance can be induced in the spin switching of magnetic nanostructures on surfaces [3], and how it can be used as a tool to investigate the magnetization dynamics of these spin systems.

When the stochastic switching of the spin states is brought into a resonant regime with an oscillating magnetic drive field or a harmonic drive voltage one gets insight into the interaction of these structures with their environment and, crucially, it enables the observation of ultrafast dynamics of excited spin states that are not easily accessible to other scanning probe techniques. This method allows for the broadband observation of spin dynamics with previously inaccessible bandwidth ranging from milliseconds to picoseconds.

We demonstrate these capabilities on magnetic nanostructures assembled with Fe atoms on a Cu$_2$N surface grown on Cu(100), but this frequency-resolved spectroscopy method should be applicable to any quantum system that can be excited by electron tunneling or force interaction with the probe tip of a scanning probe microscope.

References

Defects are frequently present in 2D materials, and as such have been extensively studied on suspended samples. However, the presence of the metallic substrates commonly employed during the growth and the characterization processes might substantially alter the electronic structure of the 2D material. In order to describe realistically the electronic properties and the SPM characterization of such samples, simulations need to take the impact of the substrate into account.

The interaction between metallic substrates and pristine transition metal dichalcogenides (TMDs) can greatly vary depending on the metal [1]. We have studied the interaction of several point-like defects in TMDs monolayers with underneath Ir(111) and Au(111) substrates by means of DFT calculations and SPM simulations, revealing a notably different behavior depending on the metallic substrate considered. The hybridization of the S states with those of the Ir(111) substrate induces a shift of ~1 eV of the MoS$_2$ states towards the valence band and a large broadening of the defect states [2]. The interaction with a gold substrate is much weaker, as confirmed by experimental data [3], leading to sharper defect states (Fig.1), much more similar to those found for freestanding MoS$_2$.

References

[3] in progress

Figure 1.
Left panel: AFM (a) and STM at -0.1 V (b) images of a S vacancy at the top layer for freestanding MoS$_2$.
Right panel: LDOS of top S atoms close (dark blue) and far (light blue) of the defect site and STM images at V = -0.1 V for a top S vacancy in (c) epitaxial MoS$_2$/Au(111) and (d) MoS$_2$/Ir(111).
Inset in d): LDOS of a top S vacancy in freestanding MoS$_2$.
Magnetic impurities on the surface of spin-orbit coupled but otherwise conventional superconductors offer the possibility to create topological phases with Majorana fermions (MFs) without having to apply an external magnetic field. In this talk I will present some of our resent results in modeling single magnetic impurities, impurity wires and islands on the surface of spin-orbit coupled superconductors. In particular, I will focus on the importance of using a self-consistent treatment of the superconductivity, which results in a local pi-shift of superconducting order parameter near the magnetic impurities. For example, I will show how MFs at wire end points strongly hybridize with in-gap Yu-Shiba-Rusinov (YSR) states, causing large oscillations in the MFs energies, which are enhanced within a self-consistent treatment. Still, by treating the MF as topological boundary mode dependent only on the effective mass gap, we can arrive at a fully parameter-free fitting of the Majorana localization length, which stays very short. I will also show that, despite the clear presence of a local pi-shift of the superconducting order parameter at magnetic impurities, it surprisingly cannot be directly measured using the Josephson effect.
Brighter but not smaller: advances in light emission using scanning probe techniques in air

Elizabeth Boer-Duchemin
Institute des Sciences Moléculaires d'Orsay (ISMO), CNRS Université Paris-Saclay, Orsay, France

While beautiful work has been carried out regarding the emission of light from the junction of a scanning tunneling microscope (STM) in ultrahigh vacuum and at low temperature, there are many advantages to working in air, that is, under ambient conditions. One such advantage is the fact that the STM/AFM head may be coupled to an inverted optical microscope. In this geometry, the emitted light may be collected below the sample (through a transparent substrate) using a high numerical-aperture aberration-corrected objective lens[1]. Not only is a higher percentage of the emitted light collected in this way, real plane and Fourier plane (or back-focal-plane) images may be formed, thus yielding information on the spatial and angular distribution of the emitted light. Such information may then be used to interpret the excitation and emission mechanisms at play.

In this talk I will present examples of the tunneling current excitation of light from plasmonic and semiconductor samples under ambient conditions. I will discuss the local, electrical excitation of individual plasmonic nanoparticles[2], of plasmonic nanoparticle arrays, and the first demonstration of the “plasmon-less” STM excitation of a transition metal dichalcogenide monolayer[3].

Figure 1: Ambient STM-excited luminescence from a TMD. (a) Schematic of the experiment. (b) Optical transmission image and (c) Optical real plane image obtained during STM excitation of the TMD monolayer. The real plane image shows that while the majority of the STM-excited excitons recombine and emit light below the STM tip, others diffuse and recombine at different “hot spots” on the flake.

References

Creating new bands in a superconductor by crafting different types of spin chains

Deung-Jang, Choi\textsuperscript{1,2,3}

\textsuperscript{1}Centro de Fisica de Materiales, CFM/MPC (CSIC-UPV/EHU), Paseo Manuel de Lardizabal 5, 20018 Donostia-San Sebastian
\textsuperscript{2}Donostia International Physics Center (DIPC), Paseo Manuel de Lardizabal 4, 20018 Donostia-San Sebastian
\textsuperscript{3}Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain

E-mail: djchoi@dipc.org

Scanning tunneling microscopy (STM) has proved to be a mature technique for the study of magnetic impurities on different substrates. Additionally, the STM allows us to manipulate atoms and assemble magnetic structures of atomic dimensions that are going to behave differently depending on their geometrical and chemical environment. We have applied our techniques to the study of magnetic spectra on atomic spin structures not only on metallic system [1] but also on superconducting surfaces [2-6] revealing the topological properties [5,6]. Recently, the introduction of impurity states in the superconducting gap has received a lot of attention. Indeed, the search of a new superconducting state called topological superconductivity is strongly based in the combination of doping classical (s-wave) superconductors with magnetic impurities that arrange spins in a chiral fashion. We present the first results of controlled single-atom manipulation to assemble a chain of Cr atoms on a Bi\textsubscript{2}Pd superconductor. Such magnetic impurities on different substrates allow us to explore many-body effects and exotic phenomena in different experimental spin systems giving an understanding on the parameters on each system.

References

Atomically resolved terahertz scanning tunnelling spectroscopy as a tool for exploring new materials

S. E. Ammerman¹, V. Jelic¹, Y. Wei¹, V. N. Breslin¹, M. Hassan¹, N. Everett¹, S. Lee¹, S. Adams¹, Q. Sun², C. Pignedoli², P. Ruffieux², R. Fasel²,³, and T. L. Cocker¹

¹Michigan State University, East Lansing, United States
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Lightwave-driven scanning tunnelling microscopy achieves exquisite spatio-temporal resolution through coherent control of tunnel currents with the oscillating field of a single-cycle light pulse. It was first demonstrated at terahertz frequencies [1], which are particularly well suited to such strong-field control [2,3]. Terahertz scanning tunnelling microscopy (THz-STM) has subsequently been used to resolve the picosecond motion of single molecules [4-6] and extreme tunnel currents through single silicon atoms [7], among other exciting recent results [3]. Thanks to its unmatched combination of ultrafast temporal resolution with atomic spatial resolution, THz-STM promises further breakthroughs, especially as a tool for exploring new materials. Yet, its unique view also necessitates a deep understanding of how THz-STM measurements relate to the underlying physics of the system, as the phenomena in question may not be visible to any other experimental technique. Here, we establish an experimental [8] and theoretical [9] framework for atomically resolved terahertz scanning tunnelling spectroscopy, which we believe will be a key modality for future studies.

References

A general description of driven dissipative systems beyond the Bloch equations

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Physical systems are, in general, subjected to interaction with their environment. In the realm of quantum systems, this gives place to dissipative dynamics where relaxation and phase decoherence emerges. Moreover, dissipation also rules the evolution of quantum systems probed by a classical sensor, as found in most experimental situations. A harmonic driving is frequently used to examine the system properties, as done in magnetic resonance [1] or in quantum stochastic resonance [3] of single atoms. An equation analogous to the macroscopic Bloch equation can be derived for the commonly found two-level system weakly coupled with the environment at high driving frequencies [3]. However, experimental features incompatible with this description appear as soon as the strength of the driving increases, while theoretically, some of the approaches done are of doubtful validity [4]. Here I present a description of the dissipative driven dynamics based on a Fourier decomposition of the weak interaction with the environment. This approach also provides a robust framework for strong driving, capable of describing magnetic resonance or quantum stochastic resonance. In contrast to more general approaches, as the non-equilibrium Keldysh Green’s functions with Floquet decomposition, the proposed method keeps a simpler description, and it can be easily compared with the standard Bloch equations.

References

Chemical stability of zigzag edges in carbon nanostructures
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Carbon nanostructures with zigzag edges exhibit unique properties with exciting potential applications. However, we show here that such nanostructures are often unstable under ambient conditions even if they display a predominantly closed-shell character. In fact, exemplified with narrow chiral graphene nanoribbons (chGNRs), we show how low pressures of pure oxygen gas readily oxidize the ribbons$^1$ in spite of their predominantly closed-shell character.$^2$ The oxidation has dramatically disruptive effects on their electronic properties, which severely jeopardizes their potential applicability. That is, the lack of stability is a barrier that must be surmounted to allow for a scalable exploitation of this type of materials. We prove the viability of chemical protection/deprotection strategies for this aim on the same chGNRs.$^3$ Upon hydrogenation, the chGNRs survive an exposure to air, after which they are easily converted back to their original structure via annealing. We also approach the problem from another angle. Determination of the most reactive sites and of the nature of the main oxidation products allowed us synthesizing a chemically stable oxidized form of the chGNRs that can be subsequently converted to the pristine hydrocarbon form via hydrogenation and annealing. These findings can be extrapolated also to other carbon nanostructures with zigzag edges and may open new doors toward their integration in devices.

References

Probing geometric and electronic properties of single-molecule junctions with a combined *operando* nearfield Raman & break-junction platform

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Gathering atomic-scale information about the geometric and electronic redox properties of individual molecules trapped in an STM break-junction is highly desirable to advance our understanding of – and ultimately rationally design and control – efficient molecular electronics devices, or (physiological) electron transfer systems in general.

In my talk, I will highlight our recent methodological advances to achieve simultaneous STM-based *operando* nearfield Raman nanoscopy and plasmon-supported break-junction experiments. These approaches allow us to gain unprecedented insights into the correlation of chemical, topographic and electronic properties of individual (bio)molecules trapped in an STM tip-substrate gap under electrochemical working conditions, *i.e.* in electrolyte and potential and/or current control. In this way, we elucidate adsorption geometry, chemical interaction and conversion processes and molecular conductance properties with extreme spatial resolution under reaction conditions.

References

Spin physics in graphene nanostructures

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Atomic-scale control over size, shape, and composition of graphene nanostructures have become a reality through on-surface synthesis whereby suitably designed chemical precursor molecules are assembled and reacted on a metal substrate under vacuum conditions. This has led to the realization of fascinating open-shell nanographenes and nanoribbons with interesting topological and magnetic properties.

In this talk I will discuss our theoretical contributions to understand and describe the electronic and spin properties in a range of graphene nanostructures [1-7]. The emergence of localized electron spins in these structures appear as promising candidates for applications in quantum technologies, provided that the interaction with their environment can be controlled. Here I will mention our efforts to characterize (i) electron spin coherence in the presence of hyperfine coupling to nuclear spins and (ii) electron beam splitting and spin transport in multi-terminal devices composed of crossed graphene nanoribbons [7].

References

**Real-space sub-femtosecond imaging of quantum electronic coherences in molecules**

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Tracking electron motion in molecules is the key to understand and control chemical transformations. Contemporary techniques in attosecond science have the capability to generate and trace the consequences of this motion in real time, but not in real space. Scanning tunnelling microscopy (STM), on the other hand, can locally probe the valence electron density in molecules, but cannot provide by itself dynamical information at this ultrafast time scale. In this talk, I will show you that by combining STM and attosecond technologies, quantum electronic coherences induced in molecules by < 6 femtosecond long carrier-envelope-phase (CEP) stable laser pulses can be directly and simultaneously visualized with angstrom-scale spatial resolution and sub-femtosecond temporal resolution. In particular, I will show that near fields of near-infrared pulses confined to the apex of a nanotip of an STM enable concurrent real-space and real-time imaging of coherences involving the highest occupied (HOMO) and lowest unoccupied (LUMO) molecular orbitals in perylenetetracarboxylic dianhydride (PTCDA) molecules, as well as exerting full control, with sub-femtosecond resolution, of the electron density in the highest occupied (HOMO) and lowest unoccupied (LUMO) molecular orbitals.

Laser pulses can couple to an STM junction either in the weak-field or in the strong-field interaction regime. The strong-field regime entails significant modification (dressing) of the tunneling barrier of the STM junction, whereas the weak field or the photon-driven regime entails perturbative interaction. In this talk, I will describe how photons carried in an ultrashort pulse interact with an STM junction, defining the basic fundamental framework of ultrafast photon-induced tunneling microscopy. Consolidation of the technique calls for innovative approaches to detect photon-induced tunneling current at the STM junction. Three techniques involving dispersion, polarization and frequency modulation of the laser pulses will be introduced to lock-in detect the laser-induced tunneling current. Photon-induced tunneling current can simultaneously achieve Ångstrom scale spatial resolution and sub-femtosecond temporal resolution. Ultrafast photon-induced tunneling microscopy will be able to directly probe electron dynamics in complex molecular systems, without the need of reconstruction techniques.
What can atomic force microscopy contribute to understand atomic-scale magnetism?

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Non-collinear magnetic structures, such as skyrmions and spin spirals, are hot candidates for nano-scale magnetic storage, and are stabilized by an interplay between competing exchange interactions. In order to understand and predict the formation of these structures, it is inevitable to quantify the different magnetic exchange interactions at the atomic scale. Spin-polarized scanning tunneling microscopy has achieved great success in investigating the magnetization of non-collinear structures at surfaces [1], but faces limitations in being able to directly detect the underlying exchange forces, as well delineate between structural and electronic contributions to the spin-polarized density of states. More than 10 years ago, a force-based method to directly probe the magnetic exchange interaction between a magnetic tip and a magnetic surface structure has been presented (magnetic exchange force microscopy) [2]. However, this method has been scarcely applied to quantify magnetic exchange forces of non-collinear magnetic structures so far.

In my talk, I will discuss what the combination of spin-polarized scanning tunneling microscopy together with magnetic exchange force microscopy (for short called SPEX) can help to sense and quantify magnetic exchange interactions at the atomic scale. I will discuss how SPEX can contribute to independently measure the geometric, electronic and magnetic structure at the atomic scale and to quantify different magnetic exchange force regimes [3]. I will show that SPEX can resolve the magnetic noncollinearity of the antiferromagnetic spin spiral in a monolayer Mn/W(110). Using distance-dependent spectroscopy together with first-principles calculations, the interplay between different magnetic exchange interactions, i.e. antiferromagnetic and Zener-type ferromagnetic exchange mechanism, can be quantified.

References
Topological superconductivity in van der Waals heterostructures

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There has been a surge of interest in designer materials that would realize electronic responses not found in naturally occurring materials. For example, it is not clear if topological superconductivity [1], which is a key ingredient in topological quantum computing, exists in any single material. These limitations can be overcome in designer van der Waals (vdW) heterostructures, where the desired physics emerges from the engineered interactions between the different components. Here, we use molecular beam epitaxy (MBE) to grow a monolayer of ferromagnetic CrBr3 on a superconducting NbSe2 substrate [2]. This combines out-of-plane ferromagnetism with Rashba spin-orbit interactions and s-wave superconductivity and allows us to realize topological superconductivity in a van der Waals heterostructure [3]. We characterize the resulting one-dimensional edge modes using low-temperature scanning tunneling microscopy (STM) and spectroscopy (STS). The use of vdW heterostructures with uniform and high-quality interfaces is promising for future device structures and further control of topological superconductivity through external stimuli (e.g. electrostatic gating).

In the second part of the talk, I am going to talk about how moire-related physics will apply to topological superconductor systems. In particular, I am going to show, using moire patterns to create a new topological region in the phase diagram at parameter values (e.g. chemical potential) where topological superconductor does not occur in the absence of the moire [4]. Our results put forward moire patterns as a powerful tool to overcome conventional constraints for topological superconductivity in vdW heterostructures. In a broader picture, periodic potential modulation provides a general way of controlling topological superconductivity towards the realization of topological qubits in the future.

References

Sifting Self-Organisation: Automated Classification of Far-From-Equilibrium Nanostructures

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Nanoparticle-solvent films deposited on solid substrates are associated with a rich dynamic behaviour that gives rise to a wide variety of striking self-organized patterns. In the far-from-equilibrium regime (i.e. where solvent evaporation is rapid), a remarkably broad array of intricate, spatially correlated patterns form including “foam-like” cellular networks, labyrinthine structures similar to those formed in spinodal decomposition of binary fluids, and fractal morphologies [1-4]. In many ways the system is a playground for self-organisation driven far from equilibrium (and, coincidentally, has many parallels with the physics of coffee stains). Its ability to generate a panoply of patterns across a wide range of length-scales provides a stringent test of the ability of machine learning algorithms to sift and classify self-organised and self-assembled structures.

We have used a combination of Monte Carlo simulations, traditional statistical approaches, and machine learning to automatically distinguish a variety of spatially correlated patterns in a broad data set of experimental AFM images of self-organized nanoparticle patterns [5]. We do this regardless of feature-scale and without the need for manually-labelled training data. I will discuss the efficacy of the machine learning approach versus more traditional statistical image analysis techniques. Although convolutional neural nets are a powerful tool, we need always be wary of Maslow’s aphorism: “If all you have is a hammer, everything looks like a nail.”

References

Tracing ultrafast hot electron dynamics inside a THz-gated STM tip

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Various approaches combining femtosecond laser excitation with scanning tunneling microscopy (STM) have been developed to image the non-equilibrium state of a sample in real-time, and new approaches such as lightwave-driven STM are currently emerging [1,2]. A major challenge for any photoexcited ultrafast STM is the unavoidable (transient) heating of the STM tip during laser illumination [3]. Specifically, femtosecond photoexcitation of the junction will generate a transient hot electron distribution inside the tip [4], which not only heats up the lattice via electron-phonon coupling, but can alter the tunneling current on femtosecond time scales and cause sample excitation by injection of hot electrons. Yet, the detailed femtosecond response of the STM tip, and in particular its electronic sub-system, is still poorly understood. Here we probe the transient hot electron gas inside a laser-excited STM tip via phase-resolved detection of single-cycle THz pulses coupled to the STM. Exact knowledge of the transient THz bias coupled to the STM [5,6] combined with numerical simulation of hot electron transmission allows us to extract the ultrafast response of the hot-electron induced current, which is directly linked to the femtosecond decay of the electronic temperature inside the tip. Whereas at large gap distances, for a nearly free-standing tip, considerable delayed emission of hot electrons is observed at high laser intensities, we find that this delayed thermionic contribution to the current vanishes in the THz response at nm gap distances. We discuss effects of the nanoscale tip geometry, optical nanolocalization, and hot electron tunneling on the electronic temperature evolution inside the tip, and propose optical field-driven tunneling as a competing channel dominating the current in the tunneling regime. Generally, our approach provides a route for probing charge carrier dynamics in photoexcited THz-STM on time scales much shorter than a THz cycle.

References

Dynamic Duo: Sparse Sampling and Parallel Spectroscopy for Fast Quasiparticle Interference Imaging

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Quasiparticle interference imaging with the scanning tunneling microscope (QPI-STM) is a crucial complement to Angle Resolved Photoemission Spectroscopy (ARPES) in the band structure characterization of quantum materials. It operates well in conditions in which other tools are limited, such as in the presence of electromagnetic fields, at ultra-low temperatures and on tiny field-effect devices. QPI-STM gathers insight into the band structure from the Fourier transform of a massive number of local density of states (LDOS) measurements, whose mapping can occupy an STM for days. Conventionally, QPI imaging works by measuring the energy dependent LDOS for every point on a grid in the field of view (FOV). While the momentum resolution improves with the physical size of the FOV, it also demands a denser sampling grid to access high-momentum states and cover the desired reciprocal space. This conundrum troubled experimentalists who had to balance momentum resolution and acquisition time. Here we introduce sparse sampling and parallel spectroscopy as two methods that accelerate QPI imaging. Although they both independently provide substantial speed-ups, their effects are fundamentally enhanced when used in combination, enabling orders of magnitude faster mapping times. Sparse sampling reduces the number of required LDOS measurements by utilizing the sparsity of the QPI pattern and parallel spectroscopy measures a spectrum faster by exploiting nonlinearities in the sample’s current-voltage characteristic. Both concepts are first introduced and then demonstrated for QPI imaging of the Au(111) Shockley surface state. We showcase three orders of magnitude faster QPI imaging, apply our fast mapping methods also on more exotic samples, and discuss further possibilities.
Dynamic Duo: Sparse Sampling and Parallel Spectroscopy for Fast Quasiparticle Interference Imaging

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Quasiparticle interference imaging with the scanning tunneling microscope (QPI-STM) is a crucial complement to Angle Resolved Photoemission Spectroscopy (ARPES) in the band structure characterization of quantum materials. It operates well in conditions in which other tools are limited, such as in the presence of electromagnetic fields, at ultra-low temperatures and on tiny field-effect devices. QPI-STM gathers insight into the band structure from the Fourier transform of a massive number of local density of states (LDOS) measurements, whose mapping can occupy an STM for days. Conventionally, QPI imaging works by measuring the energy dependent LDOS for every point on a grid in the field of view (FOV). While the momentum resolution improves with the physical size of the FOV, it also demands a denser sampling grid to access high-momentum states and cover the desired reciprocal space. This conundrum troubled experimentalists who had to balance momentum resolution and acquisition time. Here we introduce sparse sampling and parallel spectroscopy as two methods that accelerate QPI imaging. Although they both independently provide substantial speed-ups, their effects are fundamentally enhanced when used in combination, enabling orders of magnitude faster mapping times. Sparse sampling reduces the number of required LDOS measurements by utilizing the sparsity of the QPI pattern and parallel spectroscopy measures a spectrum faster by exploiting nonlinearities in the sample’s current-voltage characteristic. Both concepts are first introduced individually and then combined for QPI imaging of the Au(111) Shockley surface state. We showcase three orders of magnitude faster QPI imaging, apply our fast-mapping methods also on more exotic samples, and discuss further possibilities.

References

Imaging the effect of electron transfer at the atomic scale

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Electron transfer plays a crucial role in many chemical processes, from photosynthesis to combustion and corrosion. However, the effect of electron transfer on the electronic structure of organic molecules remains largely unclear. Unveiling these fundamental aspects requires the development of experimental tools allowing the observation of electron transfer down to the single molecule level.

In this talk, I will present an imaging approach, namely single-electron alternate-charging scanning tunneling microscopy (AC-STM), allowing mapping the orbital structure of single molecules upon electron transfer. In this way, we unveiled the effects of electron transfer and polaron formation on the single-orbital scale [1].

For the case of individual polyphenylene molecules, by combining sub-molecular-resolved atomic force microscopy with redox-state-selective orbital imaging, we characterize both structural and electronical changes occurring upon hole injection [2]. While the neutral molecule exhibits a delocalized frontier orbital, for the cationic radical the excess charge is observed to localize, inducing a partial planarization of the molecule. These results provide direct evidence for self-trapping of the excess charge in oligophenylenes, shedding light on the interplay of charge localization and structural distortion.

References

On-surface synthesis of silicene and nanographene characterized by atomic force microscopy

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Recent advances in atomically-precise nanographene surface synthesis [1] have fostered a plethora of novel functionalities in these structures such as the emergence of topological electronic states or magnetic edge states. However, proximity-induced superconductivity in the nanographene is scarce in literature [2], although it could lead to the emergence of new class of topological superconductors [3]. Extending the on-surface chemistry toolbox on superconducting surfaces is therefore a crucial step in this quest.

In this context, I will show our recent work exploring on-surface synthesis routes for silicene [4], porous GNRs [5] and Kagome graphene [6] on Ag(111) surfaces characterized by low temperature scanning tunneling microscopy (STM) and atomic force microscopy (AFM). I will also briefly discuss our first experimental attempts to induce surface reactions to superconducting substrates such as Pb(111) or Nb(100) covered with a thin Ag film [7].

References

Using SP-STM at nanometer distances for imaging and manipulation of atomic-scale magnetism

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Atomic-scale non-collinear spin textures in ultra-thin films, like magnetic nano-skyrmions, raise expectations for their application in information technology as logic spin-electronic devices or in recording media. For their characterization and manipulation spin-sensitive techniques with ultimate spatial resolution are required. Exploiting the tunneling effect between magnetic electrodes, as between a magnetic probe tip and a magnetic sample in a spin-polarized scanning tunneling microscopy (SP-STM) set up, has proven to be capable of resolving and switching magnetic structures down to the single atomic level. However, this approach disqualifies for many technical applications, since it requires tip-sample distances in the order of a few angstroms. Technical feasible distances, like flying distances of read-write heads in current data storage devices, are in the range of a few nanometers. Electron tunneling over this distance is very unlikely and therefore not detectable.

Spin-polarized resonance states (sp-RS) are unoccupied electronic states in the vacuum gap between a probe tip and a magnetic sample. As I will show in this talk, these states exhibit the same local spin quantization axis as the spin texture of the underlying sample surface, even when the spins are rotating on the atomic scale [1]. In an SP-STM setup, these states can be addressed by spin-polarized electrons that tunnel resonantly from the magnetic tip via a sp-RS into the surface, resulting in a magnetic image [1]. Our SP-STM experiments on ultrathin films with non-collinear spin textures demonstrate that this spin-polarized resonant tunneling allows for atomic-scale spin-sensitive imaging in real space at tip-sample distances of up to 8 nm, providing a loophole from the hitherto existing dilemma of losing spatial resolution when increasing the tip-sample distance in a scanning probe setup [2]. Experimental results will be discussed in terms of the sp-RS’s spin-splitting and the magnetic contrast as a function of bias and tip-sample distance, as well as in terms of the atomic-scale nature of the resonant tunneling condition.

In combination with thermally-assisted spin-transfer torque switching via sp-RS [3], our approach qualifies for a spin-sensitive read-write technique with ultimate lateral resolution, potentially opening a pathway towards future technical applications.

References

From single-molecule fluorescence to photosynthesis with an STM

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The electric current traversing the junction of a scanning tunneling microscope (STM) may lead to a local emission of light that can be used to generate sub-molecularly resolved fluorescence maps of individual molecules. Combined with spectral selection and time-correlated measurements, this hyper-resolved fluorescence microscopy approach allowed us to scrutinize the vibronic structure of individual molecules [1] in a very similar way than in the recent TERS reports, without requiring an optical excitation. We used this approach to characterize the photonics properties of charged species [2], to track the motion of hydrogen atoms within free-based phthalocyanine molecules [3], and more recently to follow resonance energy transfers between individual pigments, exploring processes occurring in photosynthetic complexes with sub-molecular spatial resolution [4]. These results constitute an important step towards photonic measurements with atoms-scale resolution [5].

Exploring energy transfers occurring in leaves with a scanning tunneling microscope.

References

From single-molecule emitters to entangled excitonic states: the insights from the scanning-probe spectromicroscopy

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Exploration of fundamental photophysical phenomena at the level of individual quantum objects requires highly specialized optical spectroscopy with ultrahigh spatial resolution, and single-photon capabilities. Recent progress in light spectromicroscopy in picocavities provided access to the excited states of molecules with such unprecedented detail. We achieve that by combining the existing 4K STM, AFM methodology with photon-detection schemes and theoretical modelling. Precisely injecting charge carriers into individual dye molecules induces formation of charged states and transient excited states (excitons), from which we then collect their emission spectra. The obtained photons carry detailed information about the energies, charges, vibronics and temporal evolution of the system state. On example cases we demonstrate how dependence of these quantities on external parameters elucidates the internal mechanisms of single emitters. Next, we address exciton delocalization over aggregates of charged chromophores with the application of extended methodology. Here, the exciton coupling mechanism is identified through an accurate determination of the aggregate geometry via AFM and by comparison of photon maps of excitonic eigenmodes with their theoretical models. Finally, we will show how control can be attained over the excitonic eigenmodes by electric field in the picocavity.

References:
In a visionary colloquium nearly sixty years ago, Richard Feynman proposed to study complex and elusive quantum systems using more controllable analogues, an approach known as quantum simulation [later published, 1]. Quantum simulators based on ultra-cold atoms, nanophotonics, trapped ions, and superconducting circuits have been developed and successfully applied to study a host of different phenomena [2]. In contrast, electronic quantum simulators have been lacking behind.

In this talk, I will demonstrate that electron gases on well-defined metal surfaces form an excellent platform for quantum simulation. By patterning the surface with atomic scale precision using a scanning tunneling microscope, the electrons can be corralled into artificial lattices of nearly any geometry. The same microscope can then be used to measure the local density of states at all positions of interest and to probe the spatial extend and shape of the electronic states. I will show several examples of how we exploit the tunability of this platform. Particular emphasis will be given to our recent efforts to create and study topological insulators.

References

Coherent Control of Individual Atomic and Molecular Spins on Surfaces

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In this talk, I will introduce recent advances achieved using ESR-STM, the combination of electron spin resonance and scanning tunneling microscopy [1]. This technique can act as a new architecture for coherent control of spins on surfaces and allows to address single atoms and molecules on surfaces with unprecedented energy resolution. Thus, it can be used to sense the magnetic coupling between spin centers on the nanoscale [2], including their dynamics [3,4]. In addition, when scanning the STM tip across the surface it permits to perform magnetic resonance imaging on the atomic scale [5]. The high energy resolution also grants access to the hyperfine interaction between the electron and nuclear spin of different atomic species [6]. Recently, we could extend this technique also to spin resonance on individual molecules [7]. Lastly, by employing pulsed ESR schemes, a coherent manipulation of the surface spin becomes possible, for instance in Rabi and Hahn echo schemes [8]. This opens up a path towards quantum information processing and quantum sensing using atomic building blocks, including atoms and molecules.

References

https://www.researchsquare.com/article/rs-134144/v1

Fig: Artistic illustration of single electron spin resonance of a surface spin.
Real-space evidence for wavelike electronic energy transfer in donor–acceptor molecular systems through quantum coherence

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Understanding intermolecular electronic energy transfer (EET) mechanism in donor-acceptor systems is very important for engineering light harvesting in photosynthesis and photovoltaics. Different from the hopping-like incoherent Förster resonance energy transfer (FRET) at relatively large intermolecular distances, the excitation energy at small distances is forecasted to delocalize over the whole donor–acceptor system and transfer in a wavelike quantum-coherent manner [1]. However, the direct experimental demonstration of its existence remains elusive up to date [2,3], due to the challenge in detecting the fragile quantum coherence in lossy molecular systems.

In this talk, I shall present the real-space investigation on the EET in artificially constructed donor–acceptor model systems using sub-nanometer-resolved scanning tunneling microscope induced electroluminescence (STML) technique. First, I shall briefly introduce the STML technique and the spectroscopic imaging features for the isolated and coupled molecules [4,5]. Then, I shall demonstrate the incoherent-to-coherent transition in the EET process in real space by constructing well-defined donor–acceptor model systems with controlled separations even beyond van der Waals contact [6]. When the donor–acceptor separation is relatively large, the EET process is found to follow the FRET mechanism. Strikingly, when this distance is very small, two new and shifted emission peaks appear with “bonding/anti-bonding”-like spectroscopic imaging patterns. Such imaging patterns, together with theoretical simulations, clearly demonstrate the occurrence of quantum-coherent EET, specifically a purely electronic coherence in the heterodimer due to the phased superposition of molecular excitonic states. Furthermore, the coherent EET channel is found to be about 3 times more efficient than the incoherent channel in a one-step transfer process, highlighting its advantage in the EET process in large molecular networks. These findings will not only substantially deepen the understanding on the intermolecular energy transfer at the nanoscale, but also enable to design and engineer light-harvesting structures and quantum light sources with high efficiencies.

References

Abstracts of Posters

(in alphabetical order)
An Algorithm for Subcycle Terahertz Scanning Tunneling Spectroscopy

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Terahertz scanning tunneling microscopy (THz-STM) enables ultrafast measurements of surfaces, single molecules, and nanostructures with simultaneous sub-picosecond temporal resolution and atomic spatial resolution. In pump-probe THz-STM experiments employing femtosecond optical pump pulses, lightwave-driven tunneling by a time-delayed THz probe pulse accesses the evolving differential conductance of the tunnel junction. However, a general theoretical approach to extract the time- and voltage-dependent differential conductance from THz-STM measurements is lacking. Here, we introduce an algorithm for pump-probe THz scanning tunneling spectroscopy (THz-STS) analysis. Our approach allows us to reliably reconstruct the tunnel junction’s differential conductance in steady-state or time-dependent scenarios from simulated THz-STS data. The algorithm achieves subcycle time resolution, which we demonstrate by retrieving dynamics faster than the bandwidth of the input THz voltage transient. Subcycle THz-STS will make lightwave-driven microscopy yet more powerful as a tool for characterizing ångström-scale ultrafast dynamics in novel materials.
Efficient THz rectification in excitonic insulator point contacts

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The implementation of ultrafast THz pump probe schemes in scanning tunneling microscopes combines sub-atomic spatial and picosecond temporal resolution. These techniques can be applied to the investigation of the ultrafast response of correlated electron materials on the atomic scale. A particularly interesting correlated system for local ultrafast dynamics investigation is the excitonic insulator candidate Ta2NiSe5 [1], in which excitons spontaneously condense into an insulating ground state with prominent response in the THz frequency domain [2]. We confirm the controlled closing of the excitonic band gap by approaching the STM tip [3], creating a highly nonlinear IV-characteristic which efficiently rectifies the injected THz pulses. We characterize this Schottky-like diode and find both bandwidth into the THz regime as well as surprisingly high efficiency. These findings give insights into the ultrafast response of a highly correlated material and supports the characterization of Ta2NiSe5 as an excitonic insulator.

References

Magnetic impurities on superconductors give rise to discrete bound states inside the superconducting gap known as Yu-Shiba-Rusinov (YSR) states. Varying the impurity-superconductor coupling induces a quantum phase transition (QPT) as the YSR state energy passes through zero. The concomitant sign change in the Josephson current – a long sought for hallmark of this QPT – has remained elusive so far. Using scanning tunneling microscopy (STM), we demonstrate such a 0 to $\pi$ transition of a Josephson junction through a YSR state as we continuously change the impurity-superconductor coupling. We detect the sign change in the Josephson current by exploiting a second transport channel as a reference in analogy to a superconducting quantum interference device (SQUID), which provides a rudimentary phase sensitivity for the STM. The change in the Josephson current through the QPT is significant and demonstrates the role of the impurity spin as well as the parity change across the QPT.
The study of electromagnetic cavities is interesting because it allows to explore light-matter interactions. If an atom/molecule is placed inside such a cavity and an electronic current is driven through the nanoscale object, it is possible to obtain light emission. Interesting photon statistics may appear, for instance so-called anti-bunching, where two individual photons are unlikely to be emitted close in time. Such single-photon sources with well-controlled photon statistics are highly desirable with applications in the field of quantum cryptography.

Junctions formed with scanning tunneling microscopes (STM) may act as plasmonic cavities and have been shown to emit single photons. However, a complete theoretical picture of single-molecule light emission is challenging and has become an active area of research in the last couple of years. Here we report on our theoretical efforts to understand the physics in model systems for single-molecule light emission based on the rate-equation approach.
Order from a mess: the growth of 5-armchair graphene nanoribbons

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The advent of on-surface chemistry under vacuum has vastly increased our capabilities to synthesize graphene nanoribbons (GNRs) with atomic precision. The synthetic strategy for most of them is similar: Ullmann coupling reaction followed by cyclodehydrogenation. Here, we provide a detailed study of the growth process for the 5-atom-wide armchair GNRs starting from dibromoperylene on Au (111).

We combine bond-resolving STM and non-contact AFM with temperature-dependent XPS and theoretical calculations. With these techniques, we show how this GNR growth departs from the conventional scenario. Instead, it follows a concerted mechanism whereby two covalent bonds are formed simultaneously, concomitant with dehydrogenation. Indeed, this novel reaction pathway is responsible for the growth of straight GNR, which is remarkable considering the initial mixture of reactant isomers with irregular metal-organic intermediates. The provided insight will help understanding the reaction mechanisms of other reactants, and serve as a guide for the design of new precursor molecules.1

References

Modeling of molecular aggregates excitonic states probed by near-field spectroscopy

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Aggregates of interacting molecules display coherently delocalized electronic excitations (excitons). The intermolecular arrangement is the key parameter determining the interactions between the chromophores that define the optical properties of the aggregate.\cite{1} Therefore tailoring the structure of aggregates, probing their optical response through optical spectroscopy and reliably modeling the underlying electronic structure at a microscopic level are all crucial aspects in order to analyze and engineer aggregates with the desired properties.\cite{2} Scanning tunneling microscopy-induced luminescence (STML) is capable of detecting light emission with submolecular resolution. In this near-field spectroscopy technique, the coupling of the excitons to the localized plasmon of the nanocavity provides information that is not available in the standard far-field measurements, e.g. spectroscopically dark states.\cite{3} In this work, we theoretically address the experimentally observed delocalized excitonic states of small aggregates formed by perylenetetracarboxylic dianhydride (PTCDA) anions.\cite{4} To this end, we specifically develop a computational framework combining quantum-chemical calculations of the molecular electronic structure with a theoretical description of the corresponding optical response in the near-field. With this approach we simulate spatial maps of the emission, directly comparable to the experiments, while offering detailed information about the excitonic wavefunctions and how they evolve from the excited states of the interacting chromophores. This new methodology provides a general toolbox that can be used for the interpretation of STML data on a wide range of molecular emitters and their aggregates.

References

The subject of our research is chiral crystallization of carbohelicenes into homochiral structures. 2D chiral crystallization of carbohelicenes on metal surfaces is of paramount importance for light sensors or for electron-spin filters. Here, we report the 2D chiral crystallization of 2,2'-bispentahelicene on Au(111) and its thermal-induced dehydrogenation studied with scanning tunneling microscopy (STM). The low coverage deposition of bis[5]helicenes on Au(111) kept at 400 K, leads to formation of heterochiral zigzag chains of the (M,M)- and (P,P)-enantiomers growing along the herringbone reconstruction pattern. In the closed-packed monolayer, both enantiomers self-assemble into racemic phase and rotational and mirror domains can be differentiated. Due to its strong sterical overcrowding in its adsorbate state, the (P,M)-meso form was not observed on the surface.

Upon annealing the substrate to approximately 670 K and subsequent cooling to 50 K, the characteristic twisted shape of the bis[5]helicenes with protrusions is no longer observed and two-dimensional homochiral domains of planar coronocoronene molecular species can be distinguished. The transformation from helical to planar chiral molecules via dehydrogenation and loss of eight hydrogen atoms was confirmed by secondary ion mass spectrometry (ToF-SIMS). Further STM measurements at 7.5 K affirmed the formation of mirror and rotational 2D homochiral conglomerate domains of S_p- and R_p-coronocoronene.
Towards Shot-Noise Spectroscopy on Monolayer Fe(Te, Se) / SrTiO₃

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The quasiparticles known as Majorana zero modes (MZMs) are particles that have the peculiar properties to be their own antiparticle and to obey non-Abelian statistics. Because of the latter, MZMs are predicted to be the building blocks of topological quantum computing. Over the past years, many signatures of what could be MZMs have been reported. However, conclusive observations are still to be made. One reason for this is the similarity of MZMs to other zero-energy states, e.g. Yu-Shiba-Rusinov states or Andreev bound states. Scanning tunnelling microscopy (STM) has already shown to be a key tool in the field of condensed matter physics because of its extreme spatial resolution and spectroscopic abilities. Via a new STM-based tool, namely shot-noise spectroscopy, we aim to provide a new point of view onto the matter of MZMs.

The platform onto which we will apply the shot-noise spectroscopy measurements, is that of single unit-cell Fe(Te, Se), grown on SrTiO₃. Signatures of MZMs have been observed in spontaneously occurring atomic line defects in Fe(Te, Se) [1]. Moreover, theoretical considerations have suggested that shot-noise measurements are able to distinguish MZMs from other zero-energy states [2]. Here, we report on the preparation of atomically flat SrTiO₃(001) surfaces and the subsequent deposition of monolayer FeSe. Additionally, an overview of the next steps towards shot-noise measurements will be presented.

References

Enhanced zero-bias conductance at the edge of monolayer CrCl$_3$ interfaced with superconducting NbSe$_2$

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Engineering atomically flat interfaces by stacking different materials represent a promising route to create exotic states of matter such as topological superconductivity$^{[1][2]}$, quantum spin liquids$^{[3]}$, unconventional superconductivity$^{[4]}$, vortex-oriented ferroelectricity$^{[5]}$, etc. Here, we use molecular beam epitaxy to create a van der Waals heterostructure comprised of a monolayer ferromagnetic semiconductor CrCl$_3$, coupled to superconducting NbSe$_2$. The structural and electronic properties of the heterostructure are investigated by low-temperature scanning tunneling microscopy and spectroscopy. Our measurements reveal an enhancement of the zero-bias conductance (ZBC) at the edge of monolayer CrCl$_3$ islands as compared to that of the bulk CrCl$_3$ and the NbSe$_2$ substrate. We study the magnetic-field dependence of this ZBC as a function of the magnetic field. We observe that the edge mode tends to fade away by progressively increasing the strength of the magnetic field, and completely disappears once the superconducting state is quenched. Overall, our results reveal an intimate link between the emergence of zero energy edge modes and proximity-induced superconductivity in CrCl$_3$.

References

Quantum phase transitions (QPTs) driven by quantum fluctuations are transitions between distinct quantum phases of matter. At present, they are poorly understood and not readily controlled. Here, scanning tunneling microscopy (STM) and noncontact atomic force microscopy (nc-AFM) are used to explore atomic scale control over quantum phase transitions between two different topological quantum states of a well-defined π-conjugated polymer [1]. The phase transition is driven by a pseudo Jahn–Teller effect that is activated above a certain polymer chain length. In addition, theoretical calculations indicate the presence of long-lasting coherent fluctuations between the polymer's two quantum phases near the phase transition, at finite temperature. This work thus presents a new way of exploring atomic-scale control over QPTs and indicates that emerging quantum criticality in the vicinity of a QPT can give rise to new states of organic matter.

References

Resolving the exciton-libron coupling in ZnPc cation

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STM-induced electroluminescence has proven to be capable of resolving electronic and vibrational transitions in single phthalocyanine molecules adsorbed on thin insulating layers. However, broadening of the peaks corresponding to the neutral excited state (>4 meV), attributed to the n-n transitions involved in the electroluminescence process [1], prohibits resolving finer structure in the spectra. In contrast, excitons of a charged molecule (trion) measured with this technique typically exhibit a peak that is one order of magnitude narrower [2,3]. Here we show that ZnPc cation performing a frustrated rotation (libration) between two potential minima on the NaCl surface emits photons with energies around 1.52 eV with very unique and fine spectral characteristics. We observe a multiple-peak spectrum with a nearly-regular energy spacing of ~1.5 meV. We explain this phenomenon by coupling between the electronic transition of the charged molecule and librons, which are quantized states that correspond to the frustrated rotation of the molecule in the potential of the surface. We were able to reproduce the shape of the measured spectrum theoretically using a harmonic-oscillator approximation of the molecular librations and the Franck-Condon factors. We have derived the parameters of the harmonic potentials from evaluation of the angular dependence of the total energy corresponding to a ZnPc molecule in the electrostatic field of the NaCl substrate.

References

Orbital-selective coupling of electron vacuum states confined through atomic engineering

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Engineering artificial lattices is a powerful approach to examine the influence of certain parameters in the formation of peculiar band-structures. Several studies relied on the confinement of the surface state of Cu(111) with CO molecules to investigate the parameter space of topological lattices [1-4]. Here, we use the Cl/Cu(100) system, for which it has been shown that Cl vacancies can be very reliably manipulated [5, 6].

We show that by arranging these vacancies into square patches we can confine field emission resonances (FERs). More precisely, FERs typically arise at high bias voltages due to a confining potential out-of-plane between tip and sample. Here, we show that the formation of vacancy patches leads to an additional in-plane confinement caused by the difference in work function between Cl and Cu. This leads to the formation of particle-in-a-box FER modes with quantum numbers that can be tuned by adjusting the size of the patches. Using these patches as building blocks, we investigate how the character of the hosted state (s-like or p-like state) influences their coupling and investigate the so-called diamond lattice, that is expected to exhibit a topological end state if the building blocks host degenerate p-like states.

References

Confinement-engineered superconductor to correlated-insulator transition in a van der Waals monolayer

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Transition metal dichalcogenides (TMDC) are a rich family of two-dimensional (2D) materials displaying a multitude of different quantum ground states with broken symmetry such as charge density waves (CDW), superconductivity, and magnetism. Among this family, NbSe₂ is a paradigmatic CDW and superconducting 2D material and it realizes Ising superconductivity at the monolayer limit. NbSe₂ has long been considered to be a metal where Coulomb repulsions play a marginal role, and the superconducting state arises from conventional electron-phonon coupling. The emergence of CDW states is usually attributed to soft-phonon modes, so that symmetry broken states are not related with strong Coulomb interactions. However, related compounds in the TMDC family such as VSe₂ and TaSe₂ are known to be strongly correlated materials with competing correlated states including magnetic Mott insulating state. Also, theoretical calculations have shown that NbSe2 is close to a Mott insulating transition to a ferromagnetic state. In this work, we experimentally demonstrate that NbSe₂ is in proximity to a correlated insulating state, by controlling the strength of the electronic interactions by quantum confinement effects. We grow a crystalline sub-monolayer (ML) NbSe₂ with a wide variety of island sizes and their relative separations. Employing low-temperature scanning tunnelling microscopy and spectroscopy, we show that for ML-NbSe₂ of size several times the coherence length, repulsive electronic interactions create a phase transition from a superconducting to a correlated insulating state. This transition is rationalized from enhanced repulsive Coulomb interactions, which dramatically change the nature of the ground state in NbSe₂. We finally showed that for correlated ML-NbSe₂ islands close to the phase transition, superconducting proximity effect strongly impacts the ground state, pushing the system through the superconductor-correlated phase boundary. Our results emphasize the role of Coulomb interactions for the emergence of both CDW and superconductivity besides the typical electron-phonon driven scenarios in 2D TMDC’s.

References

The adsorption of a magnetic adatom on a superconducting substrate perturbs the Cooper pair condensate in close proximity to the surface. The unpaired magnetic moment induces localized bound states, so-called Yu-Shiba-Rusinov (YSR) states, inside the superconducting energy gap, which can be probed by scanning tunneling spectroscopy (STS). The coupling strength between the magnetic moment of the impurity and the Cooper pairs determines the energy needed for tunneling into the YSR state.

In this work we perform STS measurements on Mn adatoms on Ag islands on Vanadium. Vanadium is very reactive and therefore oxygen assembles on the surface. This makes it difficult to investigate a possible adsorption-site dependence of the YSR energies. We passivate the surface by expitaxially grown silver monolayers. The Ag is proximitized by the superconducting substrate, and we observe YSR states for Mn on Ag/V. As the coupling of the magnetic impurity with the superconductor depends strongly on the adsorption geometry, we compare the YSR states for Mn atoms on two different crystal orientations of the Vanadium, which influences the structure of the Ag islands grown on top and hence the YSR states.
Experimental connection between Yu-Shiba-Rusinov states and the Kondo effect using numerical renormalization group theory

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Magnetic impurities on superconductors give rise to Yu-Shiba-Rusinov (YSR) states in the gap. When superconductivity is quenched, the Kondo effect manifests itself as a spectral anomaly near the Fermi level. Both phenomena can be understood quantitatively with the single impurity Anderson model (SIAM) using numerical renormalization group (NRG) theory. One prediction of this theory is that the YSR energy depends universally on the ratio between the Kondo temperature and the superconducting order parameter. Nevertheless, deviations from this universal behavior have been observed in different experiments. Using a scanning tunneling microscope, we show that for a spin 1/2 impurity at the apex of a superconducting vanadium tip, both the YSR state and the Kondo peak can be quantitatively reproduced by the NRG theory using the Ljubljana code. We discuss the universal scaling predicted by NRG theory in the context of our experimental results.

References

Spectroscopic evidence of BCS-BEC crossover in FeSe monolayer

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The Bardeen-Cooper-Schrieffer (BCS) condensation and the Bose-Einstein condensation (BEC) are the two extreme limits of the ground state of the paired fermion systems. In the BCS limit, the Cooper pairs size is much larger than average interparticle distance and the condensation occurs concurrently with the pair formation. In BEC limit, the fermions are bound tightly to form performed pairs at the pairing temperature, and then condense at a lower temperature as weakly interacting bosons. Between these two limits, there exists an intriguing regime, the BCS-BEC crossover\textsuperscript{[1-6]}, where the coherence length (pair size) $\xi$ becomes comparable to the interparticle distance ($\sim 1/k_F$, $k_F$ is the Fermi wavevector), or equivalently $\Delta/E_F \sim 1$, where $\Delta$ and $E_F$ are the superconducting gap and Fermi energy. The BCS-BEC crossover is difficult to realize in solid states. FeSe monolayer is suggested as a candidate where the Fermi energy is comparable to the superconducting gap\textsuperscript{[7]}. Here we report direct evidence of BCS-BEC crossover in FeSe monolayer using scanning tunneling microscope and spectroscopy. The Fermi energy of FeSe film is tuned by the local work function of substrate underneath. As the hole density decreases, the local density of states evolves from a BCS gap to a step-like shape. In addition, the Zeeman splitting of the quasi-particle states under perpendicular magnetic field are observed in BCS, BCS-BEC crossover and BEC regime, which is consistent with the characteristics of a condensate. FeSe monolayer is demonstrated as a unique many-body system where the Fermi energy is comparable to superconducting gap and Zeeman energy and more novel phenomena are expected.

References

Plasmon-driven single-molecule motion

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Combining scanning tunneling microscopy (STM) and spectroscopy (STS) with STM-induced light emission, we demonstrate that nanocavity plasmons injected a few nanometers away from a molecule can induce molecular motion. For this, we studied the rapid shuttling motion of zinc phthalocyanine molecules adsorbed on ultrathin NaCl films. Single-molecule luminescence spectra from molecules anchored to a step edge were compared with isolated molecules adsorbed on the free surface. We found that the azimuthal modulation of the Lamb shift is almost two times larger compared to isolated molecules. A remotely induced rapid shuttling motion of isolated molecules by plasmon-molecule coupling can explain the difference. Plasmon-induced molecular motion may open an interesting playground to bridge the nanoscopic and mesoscopic world by combining molecular machines with nanoplasmonics to control directed motion of single molecules without the need for a local probe.
Superconducting gap engineering

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Superconducting behavior is strongly affected when a single impurity or chains of magnetic atoms are placed on a superconducting surface. The superconducting gap (Δ) is modified on the atomic sites surrounding the magnetic impurity on the superconducting lattice structure. Here, we will present the results for the observation in one dimension. We use an analogy between Bogoliubov-de-Gennes (BdG) set of equations and Abrikosov-Gorkov theory in a lattice model giving rise to equations of motion for the Green's functions. This allows us to express the effect of the magnetic impurity via a self-energy that contains scattering off the impurity via its potential part (non-magnetic) and its exchange part (magnetic). From our work, we can envisage to craft superconducting and non-superconducting patterns on a material by creating structures of magnetic impurities on top of the superconductor. This will create materials that can be spatially modified, leading to an engineering of the superconducting gap.

References

On-surface synthesis and characterization of cyclo[18]carbon

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Within the family of carbon allotropes, purely sp-hybridized structures are of particular interest due to their unique structural and electronic properties. The idea to form rings from n two-fold coordinated carbon atoms (cyclo[n]carbons) has been introduced by Hoffman [1] and a synthetic route to cyclocarbons has been elaborated by Diederich et al. [2] in the 1980’s. However, although glimpses of cyclocarbons have been detected in gas phase[2,3], they could never be stabilized long enough for characterization and there has been an ongoing controversy on whether cyclocarbons are polyynic, i.e., with alternating single and triple bonds, or cumulenic with only double bonds.

Atomic force microscopy (AFM) and scanning tunneling microscopy (STM) at low temperatures allow triggering certain on-surface chemical reactions by atom manipulation [4,5] and can thus facilitate the controlled formation of highly reactive molecules on inert surfaces from more stable precursors.

Using this approach, we formed cyclo[18]carbon by decarbonylation of a C24O₆ precursor [6] as well as dehalogenation of a C18Br₆ precursor [7] on a thin layer of NaCl. By comparing high-resolution AFM images with a functionalized tip and simulated AFM images of different theoretically predicted resonance structures, we identified two possible ground state structures of cyclo[18]carbon adsorbed on bilayer NaCl [6].

References

Towards tunable atomic neural networks

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A single cobalt atom on black phosphorus exhibits bistable valencies which can be used for orbital memory [1]. These two valencies can be electrically switched and read-out using the tip of a scanning tunnelling microscope. We studied the possibility of gating orbital memory, exploring the influence of a local electric field on the valency states. Specifically, we studied the distance-dependent influence of local point charges (copper atoms) on individual cobalt atoms [2]. We found that both the impurity state energy and the stochastic behavior of an individual cobalt atom are affected in proximity to a copper atom. Furthermore, we have shown that arrays of cobalt atoms on black phosphorus exhibit a multi-well energy landscape, leading to the realization of an atomic-scale neural network, namely a Boltzmann machine [3]. These atomic neural networks exhibit a strong response to external electrical stimuli, opening the door towards autonomous learning in machine learning hardware. These studies suggest that the combination of coupling and local gating of single cobalt atoms on black phosphorus is a promising route towards creating tunable atomic neural networks.

References

Probing the Renormalization of Spin Excitation Energy in Nanographenes

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Open-shell nanographenes are an interesting group of nanomaterials, which are based on polycyclic aromatic hydrocarbons. Their non-trivial magnetic ground states can be tuned by precise tailoring of their size and structure and opens the door to all-carbon-based molecular spintronics. Even though such open-shell nanographenes have been studied theoretically for more than seventy years already, experimental studies started to increase only in the recent years, together with the on-surface synthesis techniques. They have been proven to be a powerful alternative to solution-based synthesis, which remains challenging, due to the high reactivity of open-shell nanographenes.

In combination with scanning probe techniques, the electronic and magnetic properties of such carbon-based nanomagnets can be investigated directly on the surface. However, the interaction between the molecules and the conducting substrate has recently been predicted to cause, among others, a renormalization of the spin flip excitation energy of the molecule[1].

In this work, we investigate the simplest all-carbon-based coupled-spin-system, consisting of two antiferromagnetically coupled S=1/2 spins. Scanning tunneling spectroscopy and inelastic electron tunneling spectroscopy reveal the electronic and magnetic properties of 2,2'-biphenalenyl diradicals on the Au(111) surface and on thin insulators. Utilizing the different coupling strengths between molecule and substrate, a renormalization of the spin flip excitation energy has been confirmed.

References

Magnetic impurities coupled to superconducting condensates induce sharp in-gap resonances, the so-called Yu-Shiba-Rusinov (YSR) states\cite{Heinrich}. By reducing the distance between impurities, YSR quasiparticles can interact, hybridize, and eventually form bands. Here, we scrutinize the behavior of 3d atoms coupled to niobium by scanning tunneling microscopy and spectroscopy. We demonstrate how the coupling between spins and a superconducting condensate hosting an anisotropic Fermi surface can be tuned by varying the direction and distance between the impurities. We verify the existence of long range coupling as well as the crossing through a quantum phase transition\cite{Franke}, providing a promising platform for the emergence of topological superconductivity.

References

Artificial electronic lattices are a promising tool to elucidate novel effects in the quantum world. The Scanning Tunnelling Microscope (STM) allows to build the lattices by manipulating atoms with atomic precision and probing the electronic properties. The electronic lattices consist of artificial atoms on a metallic crystal with a surface state that acts as a 2D electron gas (2DEG). The 2DEG is patterned by atomic or molecular scatterers on the surface creating artificial atoms. The versatile artificial atoms are used in lattices to study the effect of structure on electronic characteristics, thus functioning as quantum simulator.

An already known system is CO on Cu(111)\cite{1,2}, however, the system has a lacking energy resolution of 80 meV due to coupling of the surface state with bulk states. In this research, a new system is tested for an improved energy resolution. Thin Ag layers were grown on Si(111), where the surface state of Ag falls into the bandgap of Si(111) reducing the coupling to the bulk bands. Ag atoms were pulled out of the surface and successfully laterally manipulated to make corral-like artificial atoms. A quantum corral was built and showed scattering of the surface state and clear s- and p-like states. Furthermore, the constructed dimer showed bonding and anti-bonding states. Muffin-tin calculations estimated a potential height of the Ag atoms of 0.65 eV. An energy broadening of 35 meV was approximated with the FWHM. Overall, Ag atoms were effectively laterally manipulated. Artificial atoms were realized with Ag atoms on thin layers of Ag on Si(111) and showed an improved energy resolution.

Figure 1: (a) Constant-current STM image of an artificial atom (r =3.15 nm) of Ag on Ag/Si(111) at -0.7 V and 0.1 nA. (b) Averaged dI/dV spectra background-divided at the centre and below the centre (dashed-line) of the atom with the s-type state at -12 meV and the p-type state at 77 meV.

References

Real-space imaging of anisotropic charge of σ-hole by means of Kelvin probe force microscopy

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The σ-hole bonding plays a key role in supramolecular chemistry [1], including the engineering of molecular crystals or in biological macromolecular systems [2]. Despite its relevance and intensive research devoted to σ-hole bonding, the existence of the σ-hole itself was confirmed only indirectly via quantum calculations [2] or crystal structures of complexes containing σ-hole donors and electron acceptors [3]. Thus, the spatial resolution of such anisotropic charge distributions on an atom represents a long-standing experimental challenge. Here it is shown that Kelvin probe force microscopy (KPFM) [4] with a properly functionalized probe (i.e. Xe, CO), can image the anisotropic charge of the σ-hole and of quadrupolar charge of a carbon monoxide molecule. These experimental results are supported by a recently developed theoretical framework in our group for the simulation of KPFM images.

References

Spin chains in a superconductor towards topological superconductivity

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The search for Majorana bound states (MBS) in solid-state platforms has motivated an increasing number of experiments, in particular, the realization of spin chains using STM in a superconducting surface is of special interest due to the high degree of precision that this technique allows. We use Green's function formalism to simulate a superconductor with a chain of magnetic atoms on its surface. The calculations obtained with this method show good agreement with experimental results on atomic Cr chains on the $\beta$-Bi$_2$Pd superconductor \cite{1,2}. We study two types of magnetic chains with different distance between the atomic impurities and orientation with respect to the surface. The calculations allow us to distinguish different topological character between the two, showing that one of the studied chains is in a non-trivial topological phase, and thus, we expect the emergence of MBS in this system \cite{1,3}.

References


Mapping Lamb, Stark and Purcell effects at a chromophore-picocavity junction with hyper-resolved fluorescence microscopy

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Super-resolution fluorescence microscopy, or recent proposals of molecules as candidates for optically controllable quantum bits or photon sources, among others, rely on the characterization and control of properties of single molecules on the microscopic scale [1]. Here we present a correlated theoretical and experimental study of the optical properties of an individual free-base phthalocyanine molecule excited electrically by the tip of a scanning tunneling microscope [2,3]. In this highly controllable setup, an atomically sharp silver apex of the and the substrate form a plasmonic nanocavity modifying thus the local electrodynamic and electrostatic environment of the molecule. We theoretically and experimentally reveal the role of the plasmonic Purcell effect, Lamb shift, and voltage-induced DC Stark effect on the spectral position and width of the electroluminescence emission line of the molecular exciton. We demonstrate that the Stark shift is responsible for spectral shifting of the luminescence spectral line yielding ~10nm shifts and dominates over the effect due to the plasmonic Lamb shift. By analyzing the broadening of the spectra, we can map the plasmon-driven lifetime shortening of the exciton (~ps) with atomic-scale resolution, revealing thus a purely optical image of the molecule unperturbed by artefacts emerging from the excitation of the molecule by the tunneling current. These results constitute the foundation for the development of correlated optical and transport atomically resolved microscopies, but also shed light on the microscopic electrodynamic and electrostatic interactions of molecular excitons in plasmonic picocavities [4].

References

Theory of Shiba-Shiba tunneling
at the edge of a Majorana chain

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The realization of the Majorana chain \cite{1}, a 1D-chain of Yu-Shiba-Rusinov (YSR) impurity states on the surface of a superconductor, suggests that Majorana states emerging at the edges can be probed by the STM. Recently, we have developed an ideal tool to probe and manipulate the edge states of a Majorana chain. It consists of a superconducting STM tip with its own in-gap YSR state created by a magnetic impurity on the tip. With this device we have studied the sharp resonant transport between the YSR state on the tip and another YSR on the sample, and have developed its theory \cite{2}.

This presentation will expand on the theory of Shiba-Shiba tunneling and present the possible opportunities to manipulate edge states of the Majorana chain. In certain parameter regimes theory predicts that the edge state will transfer from the chain to the tip. This may provide a first step towards realizing braiding of edge states using the STM.

References

\cite{1} S. Nadj-Perge, \textit{et al.}, "Observation of Majorana fermions in ferromagnetic atomic chains on a superconductor", Science \textbf{346}, 602 (2014).
Conversion of CO$_2$ into valuable added products is one of the promising ways in order to tackle climate change. Therefore, a detailed understanding of the interaction of catalyst complexes with their environment, e.g. a surface, is crucial. Here we study and compare the growth of the model catalyst fac-Re(bpy)CO$_3$Cl (bpy = 2,2'-bipyridine) with the catalyst Re(SS-bpy)CO$_3$Cl (SS-bpy = 3,3'-disulfide-2,2'-bipyridine) on the pristine Ag(001) surface by means of low temperature scanning tunneling microscopy (STM). In contrast to the first complex, the second one is equipped with a specifically designed sulfur anchor group in its backbone. Both complexes are sublimed onto the silver substrate at a temperature of 300K and the STM measurements are carried out at 77K.

Although the catalysts are chemically closely related, we find that both complexes show dramatically different growth behaviors. Mainly, for the first complex nucleation happens at steps with an orientation along the crystal directions <110> of the substrate. Further growth of these molecules results in well-aligned decorated steps and a unique structural arrangement of monolayers (ML). In contrast, the nucleation of the second complex does not only happen at step edges but also on the free terrace. The molecular growth does not depend on the step orientation as strongly as for the first complex, but it can be observed that the steps oriented along the <110> crystal axes are neglected during the growth.

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Hyperfine Interaction and Spin Coherence times in Graphene Nanoribbons

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The possibility to localize electron spins in atomically crafted graphene nanostructures opens possibilities for applications in quantum technologies. An important property of such states is their lifetime, determined by the interaction with their environment. In this work we investigate the role of hyperfine interaction on the spin coherence times for isolated graphene nanoribbons (GNRs). Using a tight-binding model for armchair GNRs, we compute the hyperfine coupling and fidelity for the topological zero-energy states. Our results show that the hyperfine coupling to a single 13C nuclear spin in the GNR can be up to around 1 micro eV with the corresponding electron-spin coherence time of the order of microseconds. Our methods also allow to study other nuclear spin species as well as arbitrary graphene nanostructure geometries.
Picosecond ultrasonics studied by THz scanning tunneling microscopy

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Coherent acoustic phonons (CAPs) are an essential resource in studying and determining the physical properties of solids and have wide-reaching applications in acoustic devices [1]. Here we show that CAP can be generated and detected on the nanoscale by a scanning tunneling microscope. A THz pulse focused onto the STM tip induces an extremely strong out-of-plane electric field [2] that results in an ultrafast force pulse between the tip and the sample surface. On a gold thin film on mica, this force launches localized phonon wavepackets into the Au film. The phonon wavepackets propagate at the speed of longitudinal acoustic phonon and can bounce back and forth between buried Au/mica interfaces and the Au surface. The resulting coherent lattice motion, measured by electron tunneling from a time-delayed probe pulse, oscillates the surface with picometer amplitudes and frequencies ranging between tens and hundreds of GHz. The possibility of controlling pressure in solids by locally generated ultrafast acoustic phonon fields provides a new way of interacting with matters on the nanoscale.

References

\textit{$p$-Terphenyl-based thiols bound to top-layer sulphur defects on MoS$_2$/Au(111) investigated by STM}

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The combination of transition-metal dichalcogenides (TMDCs) and organic molecules into hybrid inorganic-organic systems is a field gathering much interest in recent years. The use of submonolayers of the TMDC MoS$_2$ as a decoupling layer in an STM junction is already well established and allows highly resolved $\frac{dI}{dV}$ spectra, enabling us to e. g. distinguish between different rotamers of a molecule [1]. But 2D materials also have drawbacks: Their properties are highly dependent on their local structure, because defects influence their properties severely. For instance, sulphur point defects in MoS$_2$ show a localised resonance around the Fermi energy, not present in pristine MoS$_2$.

Here we show preliminary results of anchoring the thiol-based molecule CF$_3$-3P-SH (trifluoromethyl-$p$-terphenyl-thiol) into purposely created top-layer sulphur point defects in MoS$_2$ on Au(111). One end-group of the anchored molecule is bound to the defect, allowing it to rotate around the anchoring point upon excitation with the STM tip. Electronically, the localised resonance of the sulphur point defects remains present in most cases despite saturation with the anchored molecule. It seems to be localised not only at the anchoring point but along the whole molecule. Furthermore, we observe vibronic states of the molecule inside the MoS$_2$ gap that point to strong vibrational modes.

References

Development of a compact millikelvin STM for single spin resonance

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Coherent control of single atoms and molecules on surfaces has attracted a lot of attention over the past few years aiming to create new solid-state architectures for magnetic sensing and quantum information processing. One of the preeminent techniques for such studies is the recently realized combination of electron spin resonance and scanning tunneling microscopy (ESR-STM) [1]. However, crucial properties of potential quantum spins such as the spin relaxation time T1 and the phase coherence time T2 remain often constrained by the limits in instrumentation of existing microscopes. Both T1 and T2 are affected by the proximity to the tip and substrate electrodes, which provide thermally excited electrons [2-3]. Thus, higher T1 and T2 could potentially be obtained by achieving lower temperatures, for instance using a dilution refrigerator (DR). Here, we present the design and implementation of a unique DR-STM optimized for electron spin resonance, lowering the experimental temperature to the milliKelvin range (<100 mK). Besides a low temperature, the design distinguishes itself by fast cool-down and warm up times and an overall small size of the system. A secondary ⁴He closed cooling cycle, provided by a pulse tube liquefier, allows us to maintain the base temperature without refill of liquid helium.

Moreover, since the ground state population as well as T1 was found to scale with the resonance frequency [1,2], better RF transmission at high frequencies is also desired for ESR-STM. In the compact DR-STM the RF line is optimized by an overall short total length of cables as well as using high-frequency cabling down to the STM junction. As a result, we believe that our compact DR-STM will help to improve ESR-STM paving the way for quantum information processing using single spin centers on surfaces.

References

Very weak bonds to artificial atoms formed by quantum corrals

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We explored the bonding properties of the quantum corral (a circle of 48 iron atoms placed on a copper surface) reported by Crommie et al. in 1993 [1], along with variants, as an artificial atom [2] using an atomic force microscope (AFM). The original corral geometry confines 102 electrons to 28 discrete energy states, and we found that these states can form a bond to the front atom of the AFM with an energy of about 5 millielectron volts. The measured forces are about 1/1000 of typical forces in atomically resolved AFM. The confined electrons showed covalent attraction to metal tips and Pauli repulsion to CO-terminated tips. The repulsion at close distance was evident from the response of corral states created by deliberately placing single iron atoms inside the corral. The forces scaled appropriately with a 24-atom corral.

References

Internal Stark effect of single-molecule fluorescence

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The optical properties of chromophores can be efficiently tuned by electrostatic fields generated in their close environment, a phenomenon known as internal Stark effect (ISE) that plays a central role for the optimization of complex functions in living organisms. Here, we realised an ISE experiment at the lowest possible scale, by monitoring the Stark shift generated by integer charges confined a single chromophore on its emission energy. To this end, a scanning tunneling microscope (STM) functioning at cryogenic temperatures is used to sequentially remove the two central protons of a free-base phthalocyanine chromophore deposited on a NaCl-covered Ag(111) surface. STM-induced fluorescence measurements reveal spectral shifts that are associated to the electrostatic field generated by the internal charges remaining in the chromophores upon deprotonation.
Long-lived spin excitations of a magnetic molecule on a proximitized superconductor

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Magnetic molecules adsorbed on surfaces have been used as a platform to individually address and manipulate spins. Long spin-relaxation times are required in order to be able to use atomic spins in quantum information processing and data storage. Normally, coupling of the spin with the conduction electrons of metallic substrates can quench the excited state lifetime and lead to short relaxation times, but the presence of superconducting paring effects in the metal substrate can protect the excited spin from relaxation [1]. Here, we use a substrate of a few monolayers of gold epitaxially grown ontop of an oxygen reconstructed 1x5-V(100) surface to decouple the molecular spin of an iron-porphyrin-chloride from itinerant electrons. The gold film exhibits a proximitized superconducting gap, which protects molecular spin excited states and results into a lifetime of $\tau=100$ns. We further show that the coupling between the molecule and the substrate can be increased by the scanning tunnelling microscope tip, which results in the emergence of YSR states within the superconducting gap.

References

Free coherent evolution of a coupled atomic spin system initialized by electron scattering

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Full insight into the dynamics of a coupled quantum system depends on the ability to follow the effect of a local excitation in real-time. Here, we trace the free coherent evolution of a pair of coupled atomic spins by means of scanning tunneling microscopy. Rather than using microwave pulses, we use a direct-current pump-probe scheme to detect the local magnetization after a current-induced excitation performed on one of the spins. By making use of magnetic interaction with the probe tip, we are able to tune the relative precession of the spins. We show that only if their Larmor frequencies match, the two spins can entangle, causing angular momentum to be swapped back and forth. These results provide insight into the locality of electron spin scattering and set the stage for controlled migration of a quantum state through an extended spin lattice.
Synthesis and Characterization of Nitrogen-Doped Triangulene on Metal Surfaces

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Hydrocarbon molecules with intrinsic magnetic properties were theoretically devised already in the 1950’s, like the case of triangulene. However, its synthesis has only been successfully achieved in recent years [1]. Including a nitrogen heteroatom into its backbone may allow adjusting its magnetic state [2], but the molecular synthesis poses again serious challenges. Herein, we report the successful synthesis N-triangulene on the Au(111) and Ag(111) surfaces, along with their characterization by scanning tunneling microscopy and spectroscopy in combination with density functional theory calculations. N-triangulenes were obtained by reducing ketone-functionalized N-triangulene molecules with atomic hydrogen, followed by thermal annealing and, if necessary, manipulations with the scanning probe. We demonstrate that on Au(111) N-triangulene donates an electron to the substrate and exhibits an open-shell triplet ground state. This is derived from the different Kondo resonances of the final product N-triangulene and a series of intermediate products on Au(111), further confirmed by the excellent match between experimentally mapped and density functional theory (DFT) calculated molecular orbitals of a positively charged N-triangulene. In contrast, N-triangulene on Ag(111) receives an extra electron from the substrate and displays a closed-shell character. Our study reveals the electronic properties of N-triangulene on different metal surfaces and offers an efficient approach for the fabrication of reactive open-shell nanostructures on surfaces.

References

Line shape analysis of the eigenstates of an artificial atom formed by a quantum corral

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Surface state electrons on a Cu(111) surface can be scattered by adsorbates [1]. Manipulation of these scatterers opens the path to a variety of artificial potential landscapes. In 1993 Crommie et al. [2] assembled 48 Fe adatoms in a circle with a diameter of 15 nm. This so-called quantum corral confines the surface electrons within a circularly symmetric potential well. Past investigations with scanning tunneling microscopy and tunneling spectroscopy revealed discrete, Bessel-type eigenstates of the enclosed electrons. But these studies on the Fe-quantum corral showed unwanted movement of the corral walls during spectroscopic measurements.

[2]

Instead of Fe, we used CO molecules to upgrade the quantum corral with more stable walls. This permitted us to access a larger voltage window and allowed for a detailed line shape analysis of the resonant eigenenergy levels. The motivation of this work is to determine the lifetime of confined surface state electrons on a metal surface before investigating a corral on a topological insulator. [2]

References

Open Quantum System Model of Electron-Electron Double Resonance of Engineered Atomic Structures on a Surface in an STM

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The combination of the scanning tunneling microscope with electron spin resonance spectroscopy (ESR-STM) has opened a new dimension in the characterization of electronic surface states at the nanoscale.[1] In this work I will present our recent development [2] of electron-electron double resonance of a coupled S=1/2 dimer in an ESR-STM using a novel driving scheme that allows to drive ESR on a spin that is not directly in the tunnel junction.

I will focus on presenting a theoretical model of an open quantum system that we used to extract lifetimes (T₁ times) of the system. We find excellent agreement between simulation and experiment for a very narrow selection of parameters. In particular, I would like to highlight the significantly enhanced lifetime of the spin that is not subject to the tunnel current which reaches up to 150 ns – long enough to perform several Rabi oscillations with properly chosen driving strength. Our finding imply that this system is suitable for quantum coherent manipulation of multiple coupled surface spins, the goal of quantum coherent nanoscience.

References

Energy dissipation in nc-AFM at incipient ferroelectric perovskites

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Energy dissipation measurements in the frequency-modulated non-contact atomic force microscopy (nc-AFM) help to reveal subtle phenomena at semiconductor surfaces. The excitation channel is particularly useful for the disentanglement of various possible sources of electromagnetic interactions between a tip and sample. Here we present the potential of using dissipation as a fingerprint of polarization at incipient ferroelectric perovskites SrTiO₃ and KTaO₃ [1],[2]. Those materials, although being quantum paraelectric, can spontaneously polarize upon extrinsic doping, strain, or due to surface polarity.

In this poster bias-dependent excitation peaks will be presented for bulk-terminated SrTiO₃(100), KTaO₃(100), CO-adsorbed SrTiO₃(100), water reconstructed KTaO₃(100), and Nb-doped KTaO₃(100). I will discuss how the energy dissipates on polarized ferroelectric domains and what can be learnt from it. Additionally, I will show the capabilities of Kelvin Probe Force Microscopy (KPFM) for the polarization characterization at the nanoscale. All experiments were performed using qPlus STM/AFM sensors at UHV conditions and temperature ranging from 4K to 30K, which allowed to determine not only the bias and distance dependence of dissipation but also the effect of approaching the transition temperature.

References

Investigation of the Electronic Properties of 2D Materials Using Accelerated Quasiparticle Interference Mapping

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Along with angle-resolved photoemission spectroscopy (ARPES), quasiparticle interference (QPI) mapping using a scanning tunneling microscope (STM) have been the prevalent techniques in the characterization of the band structure of 2D systems. The latter technique is particularly suited to probe the local scattering properties of quasiparticles and dispersion relation of occupied and unoccupied states. Additionally, QPI enables the exploration of the electronic properties of surfaces in an extended parameter space such as magnetic field, ultra-low temperatures and on small devices. Although QPI mapping is an important tool for such investigations, it has the downside of being slow. This fact stems from involving two serial tasks that are performed with the tip of an STM: 1. visiting different locations on the respective surface and ramping the voltage during bias spectroscopy. In order to speed-up things, we introduce the intuitive solution: “Measure less, faster” by demonstrating the implementation of sparse sampling and parallel spectroscopy to QPI mapping\([1]\). In particular, the combination of the two techniques improves the mapping time of QPI imaging by orders of magnitude. We introduce our methods by investigating Au(111) due to its well-studied electronic properties and we explore the applicability of our methods on a more exotic system, BSCCO. In both cases, we find QPI maps in very good agreement with previous works. We show the non-invasive and reversible upgrade of an STM, which reduces mapping time from days to minutes.

References

\[\text{[1] Zengin et al., arXiv:2102.00054, 2021}\]
Spin torque in a Josephson junction between two superconducting magnetic impurity states

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Superconducting tunneling between spin-polarized Yu-Shiba-Rusinov (YSR) impurity states can be realized using a functionalized mK-STM \cite{Huang2020}, which can be further developed as a local probe of electronic spins for spintronics applications. Here, we consider a Josephson junction containing two magnetic impurities and show that the Josephson current is spin-dependent and accompanied by a spin torque. The torque acts to align the two impurities either parallel or anti-parallel, depending on the parity of YSR state's occupation.

Using standard Green's functions techniques, we derive the spin-torque and spin-current as function of the superconducting phase difference and the relative angle between the impurity spins, modeled as classical magnets. Our results are also relevant for recent realizations of double quantum dot superconducting junctions with YSR states \cite{Saldana2018}. Finally, we provide a discussion on spin dynamics with a possible relevance to spin chains that show topological superconductivity.

References

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