

Plenty of Room at the Bottom – New Developments in Scanning Probe Tools

801. WE-Heraeus-Seminar

13 Nov - 17 Nov 2023

at the Physikzentrum Bad Honnef, Germany

The WE-Heraeus Foundation supports research and education in science, especially in physics.
The Foundation is Germany's most important private institution funding physics.

**WILHELM UND ELSE
HERAEUS-STIFTUNG**



Introduction

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 801. WE-Heraeus-Seminar:

Scanning Probe Microscopy constitutes one of the most powerful set of experimental techniques to enter the atomic scale. Since their invention in the 1980s both the scanning tunnelling microscope (STM) and the atomic force microscope (AFM) were improved and are now established as atomic "workbenches" that can be used to interact with atomic-scale objects, such as molecules and single atoms. It moreover allows to characterize new phases of matter atom-by-atom – at cryogenic temperatures close to absolute zero temperature and high magnetic fields. The prospects of this field and the advent of its development was captured in Richard Feynman's famous quote and respective speech entitled "There is plenty of room at the bottom", which gives this seminar its name.

The goal of this WE-Heraeus seminar is to present the recent progress in this exciting field. This should happen with a strong focus on involving and educating the young researchers in the community. In order to do that the invited speakers from Germany, Europe and abroad will represent the excellent advances made in their respective field. The talks are grouped into 5 topical sessions, that showed outstanding progress in the last couple of years. The speakers come from both experiments as well as theory (with a strong focus on experimental speakers). Poster sessions will give in particular master students, PhD students and early-stage postdocs the opportunity to present their work to the community.

Scientific Organizers:

Dr. Deung-Jang Choi

Centro de Física de Materiales, Spain
E-mail: deungjang.choi@ehu.es

Dr. Laerte Patera

University of Innsbruck, Austria
E-mail: Laerte.Patera@uibk.ac.at

Dr. Philip Willke

Karlsruhe Institute of Technology, Germany
E-mail: philip.willke@kit.edu

Introduction

Administrative Organization:

Dr. Stefan Jorda
Martina Albert

Wilhelm und Else Heraeus-Stiftung
Kurt-Blaum-Platz 1
63450 Hanau, Germany

Phone +49 6181 92325-14
Fax +49 6181 92325-15
E-mail albert@we-heraeus-stiftung.de
Internet: www.we-heraeus-stiftung.de

Venue:

Physikzentrum
Hauptstrasse 5
53604 Bad Honnef, Germany

Conference Phone +49 2224 9010-120

Phone +49 2224 9010-113 or -114 or -117
Fax +49 2224 9010-130
E-mail gomer@pbh.de
Internet www.pbh.de

Taxi Phone +49 2224 2222

Registration:

Martina Albert (WE-Heraeus Foundation)
at the Physikzentrum, reception office
Monday (17:00 h – 21:00 h)
and Tuesday (08:00 – 12:30 h)

Program

Program (CET)

Monday, 13 November 2023

17:00 – 21:00	Registration	
From 17:45	<i>BUFFET SUPPER</i>	
19:30 – 19:45	Scientific organizers	Opening and Welcome
19:45 – 20:00	Stefan Jorda	About the Wilhelm and Else Heraeus Foundation
20:00 – 21:00	Keynote 1 Sander Otte	Atomic spins on surfaces: from early investigation to coherent control

Tuesday, 14 November 2023

08:00	<i>BREAKFAST</i>	
09:00 – 09:45	Yujeong Bae	Quantum Control of Artificial Spin Structures Built Atom by Atom on surfaces
09:45 – 10:30	Nicolas Lorente	Theory of single-impurity electron spin resonance on surfaces
10:30 – 11:00	<i>COFFEE BREAK</i>	
11:00 – 11:45	Jose Lado	Hamiltonian learning and triplon quantum matter in atomically assembled spin lattices
11:45 – 12:30	Poster Flash	
12:30 – 12:35	Conference photo	

Program (CET)

Tuesday, 14 November 2023

12:35 – 14:00	<i>LUNCH</i>	
14:00 – 15:30	Poster Session I	
15:30 – 16:00	<i>COFFEE BREAK</i>	
16:00 – 16:45	Manuel Gruber	Molecular Magnetism on Surfaces
16:45 – 17:15	Lorenzo Poggini	Quantum features of magnetic molecules on surfaces: from “innocent” surfaces to type I superconductors
17:15 – 18:00	Christian Ast	Superconducting Quantum Interference at the Atomic Scale
18:00 – 20:00	<i>DINNER</i>	
20:00 – 21:00	Keynote 2 Sabine Maier	On-surface synthesis: A bottom up strategy to atomically precise materials

Program (CET)

Wednesday, 15 November 2023

08:00	<i>BREAKFAST</i>	
09:00 – 09:45	Alexander Riss	Measuring Chemical Reactivity at the Atomic Scale
09:45 – 10:30	Oliver Gröning	Engineering of robust topological quantum phases in graphene-derived nanostructures
10:30 – 11:00	<i>COFFEE BREAK</i>	
11:00 – 11:45	Anna Rosławska	Light-matter interaction and photochemistry probed with sub-molecular precision
11:45 – 12:30	Daniel Ebeling	Chemical bond imaging: A tool to decipher on-surface reaction processes and construct organic nanomaterials molecule by molecule
12:30 – 14:00	<i>LUNCH</i>	
14:00 – 18:00	Excursion	
18:30	HERAEUS DINNER	
		<i>(social event with cold & warm buffet and complimentary drinks)</i>

Program (CET)

Thursday, 16 November 2023

08:00	<i>BREAKFAST</i>	
09:00 – 09:45	Paolo Sessi	Interfacing magnetism with superconductivity: visualizing interactions from 0d to 2d
09:45 – 10:00	Benjamin Mallada	Visualization of π-hole in molecules by means of Kelvin probe force microscopy
10:00 – 10:15	Wantong Huang	Spin engineering in artificial atom-molecule hybrids
10:15 – 10:30	Song Jiang	A Universal Model for STM-Induced Luminescence
10:30 – 11:00	<i>COFFEE BREAK</i>	
11:00 – 11:45	Franz Giessibl	Exploring the femtoscale in forces and distances
11:45 – 12:30	Shadi Fatayer	Investigating molecular properties through charge-state control
12:30 – 14:00	<i>LUNCH</i>	
14:00 – 15:30	Poster Session II	
15:30 – 16:00	<i>COFFEE BREAK</i>	
16:00 – 16:45	Martin Setvin	Imaging and Tracking Single Polarons by STM/AFM
16:45 – 17:15	Angelika Kühnle	Desorption from surfaces: Impact of molecular-level details on desorption kinetics
17:15 – 18:00	Aparajita Singha	Quantum sensing with single spin sensitivity
18:00 – 20:00	<i>DINNER</i>	
20:00 – 21:00	Keynote 3 Joseph A. Stroscio	Unraveling Orbital Magnetism Contributions to Landau Levels in Moiré Quantum Matter

Program (CET)

Friday, 17 November 2023

08:00	<i>BREAKFAST</i>	
09:00 – 09:45	Bruno Schuler	Cross-Correlation Microscopy of Atomic Defects in in Transition Metal Dichalcogenides
09:45 – 10:30	Markus Morgenstern	Scanning tunneling spectroscopy of 2D materials: Quantum Hall edge states and topological magnetic gaps
10:30 – 11:00	<i>COFFEE BREAK</i>	
11:00 – 11:45	Felix Lüpke	Artificial quantum states in assembled van der Waals heterostructures studied by scanning tunneling microscopy
11:45 – 12:30	Christian Lotze	From high-resolution molecular spectroscopy to moiré-tuned spin interactions of individual atoms: MoS₂/Au(111) as a versatile lab bench for STM studies
12:30 – 12:45	Scientific organizers	Poster Prize Awards & Closing Remarks
12:45 – 14:00	<i>LUNCH</i>	

End of the seminar and departure

Posters

Posters

- Aji Alexander **Single-atom co-catalyst dispersion on perovskite surfaces by polarity compensation**
- Jonas Allerbeck **Efficient and continuous carrier-envelope phase control for terahertz lightwave-driven scanning tunneling microscopy**
- Busra Gamze Arslan **Intricacies in 1T-TaSe₂ Islands with Deposited Cobalt Atoms, A Spinon Kondo Effect?**
- Laric Bobzien **Near-Field Amplitude Calibration for THz-STM of Atomic Defects in 2D Semiconductors**
- Alice Bremerich **Electronic Structure of Quasi-Freestanding WS₂/MoS₂ Heterostructures**
- Hong Bui Thi **Electric-field-driven Spin Resonance by an Exchange-coupled Single-atom Magnet**
- Mark Canavan **Coherent Spin Dynamics Between an Electron and Nucleus Within a Single Atom**
- Verena Caspari **Shot-noise measurements of single-atom junctions using a scanning tunneling microscope**
- Caiyun Chen **Visualizing the Localized Electrons of a Kagome Flat Band**
- Fábio Costa **Nanoscale characterization and light emission aspects of defects in h-BN**
- Jan Cuperus **Electronic characterization of magnetic CrCl₃ monolayers on NbSe₂**

Posters

Pranjit Das	Tuning the planarity of a thiantherene-based molecule on Au(111)
Adrian Ebert	Interaction of single 4f metal-organic molecules with light
Eugenio Gambari	A scanning tunnelling microscope combined with radio frequency to detect magnetic resonance at the atomic scale
Lukas Gerhard	An electrical molecular motor driven by angular momentum transfer
Paul Greule	Development of a closed-cycle dilution refrigerator scanning tunneling microscope
Caroline Hommel	Sample preparation in UHV for STM measurements
Longfeng Huang	Reactivity of single metal atoms in different coordination environments
Wantong Huang	Spin engineering in artificial atom molecule hybrids
Xin Huang	Rendering a doped Mott insulator in a NbSe ₂ heterostructure
Song Jiang	A Universal Model for STM-Induced Luminescence
Keda Jin	Creation of graphene quantum dot nanoarrays in van der Waals heterostructure
Liwei Jing	Strain-Induced Edge States on Bilayer SnTe

Posters

Divya Jyoti	Quantum Phase transitions of spin-chains on superconductors
Kyungmin Kim	Scanning Near-Field Optical Microscopy study of Metal-Insulator Transition dynamics in Vanadium Dioxide thin film
Annique Krieg	Quantum simulator to emulate lower dimensional physics and chemistry
Vivek Krishnakumar Rajathilakam	Measuring Yu-Shiba-Rusinov state dynamics using Stochastic Resonance Spectroscopy
Hugo Le Du	Huge doping and unconventional superconductivity in NbSe ₂ misfit compounds
Jinwon Lee	Signatures of an amorphous liquid of impurity states in FeTe _{0.55} Se _{0.45} superconductor
Kurt Lichtenberg	Ultrafast measurements of Charge Density Wave dynamics with a THz light-coupled STM
Leonard-Alexander Lieske	Distance-Dependence of Orbital Density Mapping Using a CO-Functionalized STM Tip
Rian Ligthart	Towards topological edge states with artificial electronic lattices: Cs on InAs(111)
Zhengyuan Liu	Aging behavior in a self-induced spin glass
Sandip Maity	Development of an Ultra High Vacuum and Low Temperature Scanning NV Magnetometer

Posters

- | | |
|---------------------------|--|
| Benjamin Mallada | Visualization of π -hole in molecules by means of Kelvin probe force microscopy |
| Luis Enrique Parra López | Probing the coherent amplitude mode in 1T-TaS ₂ with THz scanning tunneling microscopy |
| Fabian Paschke | Fusing Open-Shell Molecules on NaCl by Atom Manipulation |
| Ricardo Javier Peña Román | Investigating Spin Noise and Stray Magnetic Field in Synthetic Antiferromagnetic Structures using Nitrogen-Vacancy Scanning Probe Microscopy |
| Jose Reina Galvez | Modulation of the tunneling barrier in an Anderson impurity Hamiltonian to study ESR-STM |
| Daniel Rothhardt | Atomically thin transition metal halide NiBr ₂ grown on Au(111) |
| Lisa Rütten | Controlling hybridization in Yu-Shiba-Rusinov dimers through a charge-density wave |
| Paul Philip Schmidt | Tip induced ring growth of Pb-islands on Si(111)-(7x7) |
| Johannes Schust | Atomically resolved luminescence in a Terahertz Scanning Tunneling Microscope |
| Johannes Schwenk | Probing the magnetism of Tb atoms on MgO |
| Lisanne Sellies | Excited-state spectroscopy of individual molecules with atomic force microscopy |

Posters

Jeongah Seo	Defect identification of Nitrogen-doped graphene on Pt (111) using AFM/STM
Shixuan Shan	Enhancing ESR-STM Measurements with Python Instrument Control Library
Junyoung Sim	Coupling Single-Cycle Terahertz Fields into a Scanning Tunneling Microscope: Characterization and Time-Resolved Measurements
Evert Stolte	STM method to measure the lifetime of a single nuclear spin
Marco Thaler	Local Electronic Structure of h-BN on Pt(110)
Auke Vlasblom	On the Edge State of Finite 2D Bi₂Se₃ Crystals
Tobias Wichmann	Phonon gap supported tunneling and Faraday screening through graphene

Abstracts of Lectures

(in alphabetical order)

Quantum Control of Artificial Spin Structures Built Atom by Atom on surfaces

Yujeong Bae^{1,2}

¹Center for Quantum Nanoscience, Institute for Basic Science, Seoul 03760, Republic of Korea

²Department of Physics, Ewha Womans University, Seoul 03760, Republic of Korea

Email (Presenter): bae.yujeong@qns.science

The last few decades have witnessed remarkable revolutions of quantum mechanics and information science, driven by the desire to employ quantum states for carrying and processing information. While various material platforms have been extensively studied for realizing quantum technologies, single atoms and molecules on surfaces have been recently explored for demonstrating quantum information processing and quantum sensing since the successful integration of electron spin resonance techniques with scanning tunnelling microscopy (ESR-STM) [1,2].

In this talk, we will focus on recent advancements on quantum-coherent experiments performed using ESR-STM that enables coherent control of spins at the Angstrom length scale. Since the first demonstration of coherent manipulation of single spins on surfaces [3], one of the biggest challenges has been extending this study to multiple spins, as STM studies are typically localized to the spin located at the STM junction. Recently, we have developed a way to control and detect spins that are not positioned at the STM junction. Based on this new approach, we successfully demonstrated the coherent manipulation of multiple $3d$ -electron qubits built on a surface [Yu]. This work exhibits the enhanced coherent properties of remote spins as well as rapid controlled operations of multi-electrons in an all-electrical fashion. In addition, we will present our recent efforts to expand this work to the $4f$ electron systems [4], where we anticipate prolonged spin relaxation and coherence times due to the intrinsic isolation of $4f$ electrons. Lastly, we will discuss atomic-scale quantum sensing achieved using ESR-STM with a spin sensor attached to the STM tip, which enables detection of magnetic and electrostatic fields at subnanometer precision. This series of recent advancements represents the versatile applications of ESR-STM in the realm of quantum technologies.

References

- [1] S. Baumann, W. Paul, T. Choi, C. P. Lutz, A. Ardavan, and A. Heinrich. (2015) Electron paramagnetic resonance of individual atoms on a surface. *Science* 350(6259), 417.
- [2] Y. Chen, Y. Bae, and A. Heinrich. (2022) Harnessing the quantum behavior of spins on surfaces. *Adv. Mater.* 2107534
- [3] K. Yang, W. Paul, S. Phark, P. Willke, Y. Bae, T. Choi, T. Esat, A. Ardavan, A. J. Heinrich, and C. P. Lutz. (2019) *Science* 366(6464) 509
- [4] S. Reale, J. Hwang, J. Oh, H. Brune, A. J. Heinrich, F. Donati, and Y. Bae. (2023) arXiv. K. Yang, W. Paul, S. Phark, P. Willke, Y. Bae, T. Choi, T. Esat, A. Ardavan, A. J. Heinrich, and C. P. Lutz. (2019) *Science* 366(6464) 509

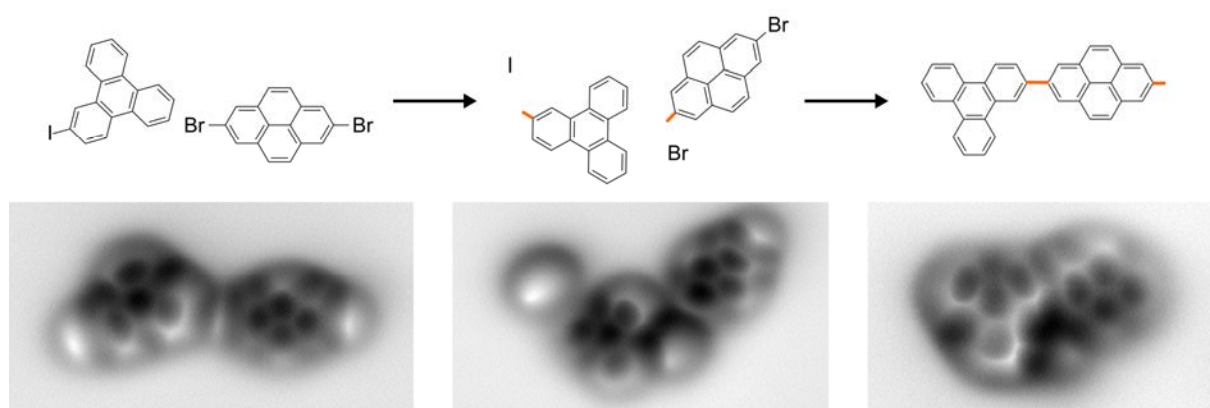
Chemical bond imaging: A tool to decipher on-surface reaction processes and construct organic nanomaterials molecule by molecule

Daniel Ebeling^{1,2}

¹Institute of Applied Physics, Justus Liebig University Giessen, Germany

²Center for Materials Research, Justus Liebig University Giessen, Germany

Constructing low-dimensional covalent assemblies with tailored size and connectivity is the key for applications in molecular electronics where optical and electronic properties of the quantum materials are highly structure dependent. We present a versatile approach for building such structures molecule by molecule on bilayer sodium chloride (NaCl) films with the tip of an atomic force microscope, while tracking the structural changes with single-bond resolution. [1] Covalent homo-dimers in *cis* and *trans* configurations and homo-/hetero-trimers were selectively synthesized by a sequence of dehalogenation, translational manipulation and intermolecular coupling of halogenated precursors (see Figure). Using scanning probe manipulation, we were also able to synthesize the structurally elusive molecule P₃N₃, an inorganic aromatic benzene analogue, which is difficult to obtain via traditional synthetic methods due to its high reactivity. [2] This work shows ways for synthesizing elusive molecules and organic nanoarchitectures, studying structural modifications and revealing pathways of intermolecular reactions. Moreover, possibilities for tweaking the bond imaging technique using torsional and flexural higher eigenmodes of qPlus sensors will be illustrated. [3] Torsional eigenmodes can be utilized to perform lateral force microscopy with single bond resolution.



[1] Q. Zhong et al. Nature Chemistry **13**, 1133 (2021)

[2] Q. Zhong et al. Angew. Chem. Int. Ed. e202310121 (2023)

[3] D. Martin-Jimenez et al. Nanoscale **14**, 5329 (2022)

Investigating molecular properties through charge-state control

S. Fatayer¹

¹*Applied Physics Program, King Abdullah University of Science and Technology, Thuwal, Saudi Arabia*

The physicochemical properties of molecules adsorbed on surfaces are charge-state dependent. Insulating films serve as an ideal platform to study the physics and chemistry of charged molecules, because these films avoid charge leakage. However, investigating the properties of individual charged molecules on insulators is experimentally challenging. The atomic force microscope (AFM), capable of single-electron sensitivity [1] and atomic resolution [2], is suitable to be used with insulators. Here I will show structural and electronic properties of molecules that can be achieved by employing AFM on insulating films – how to use the AFM as a current meter to perform tunnelling spectroscopy on molecules [3], how to perform chemical reactions with individual charges [4] and insights gained by atomically resolved AFM images of molecules with the same charge state [5].

References

- [1] L. Gross, et. al *Science* **325**, 1110 (2009)
- [2] L. Gross, et. al *Science* **324**, 1428 (2009)
- [3] S. Fatayer, et. al *Nature Nanotechnology* **13**, 142 (2018)
- [4] S. Fatayer, et. al *Physical Review Letters* **121**, 226101 (2018)
- [5] S. Fatayer, et. al *Science* **365**, 142 (2019)

Exploring the femtoscale in forces and distances

Franz J. Giessibl

Department of Physics, University of Regensburg, Germany, E-mail: franz.giessibl@ur.de

Scanning probe microscopy has brought us experimental access to the world of single atoms. The atomic force microscope (AFM), offspring of the scanning tunneling microscope (STM), has rapidly found wider applications than the STM because it allows to image any sample without requiring electrical conduction. However, the spatial resolution of AFM initially was inferior to the one of the STM. In the last years, the AFM's resolution has been boosted far beyond STM, e.g. when imaging organic molecules and even revealing electron clouds within single atoms. Height resolution reaches sub-picometer values [1], and Fig. 1 shows force contrasts beyond piconewtons [2,3]. The origin of the forces covers chemical bonding forces, Pauli repulsion forces as well as exchange forces.

Is this the end of the development of AFM? The answer is a clear no, as there are some inspiring challenges that require sizable future developmental efforts.

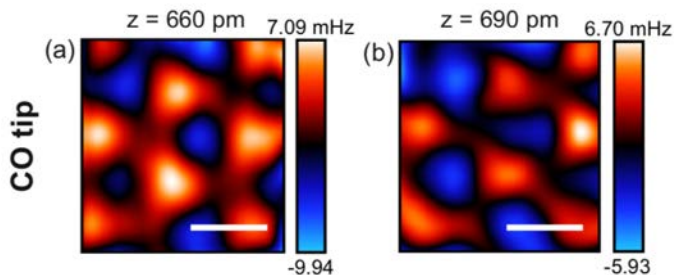


Fig. 1: Constant height AFM image of CaF_2 at core-core distance of 660 pm (a) and 690 pm (b) showing a force contrast of merely 50 fN. Adapted from [2].

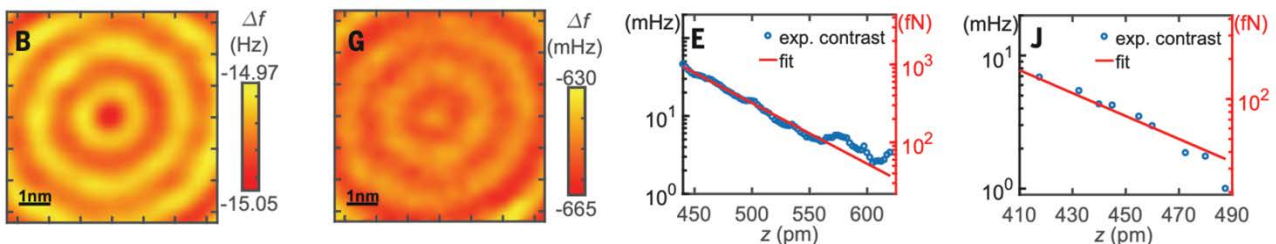


Fig. 2: Constant height image of the eigenstates in a quantum corral imaged by a metal (B,E) and CO tip (G,J) showing a force contrast of less than 100 fN. Adapted from [3].

References

- [1] F. Pielmeier, F. J. Giessibl, *Phys. Rev. Lett.* **110**, 266101 (2013)
- [2] A. Liebig, A. Peronio, D. Meuer, A.J. Weymouth, F.J. Giessibl, *New J. Phys.* **22** 063040 (2020).
- [3] F. Stilp, A. Berezuk, J. Berwanger, N. Mundigl, K. Richter, F.J. Giessibl, *Science* **372**, 1196 (2021)

Engineering of robust topological quantum phases in graphene-derived nanostructures

Oliver Gröning¹

¹*Empa Materials Science and Technology, Überlandstrasse 129, CH-8600 Dübendorf, Switzerland*

Graphene provides an ideal platform to tune its electronic properties by rational control of its nanoscale structure. Quantum confinement effects in graphene nanoribbons for example can be exploited to tune their electronic band gap and specific edge and hetero-junction topologies can lead to localized in-gap states showing π -electron magnetism. However, atomically precise synthesis of these graphene-derived nanostructures is the key to fully control their electronic properties and achieve desired functionalities. In this context, on-surface synthesis together with scanning probe techniques and atomistic simulation, e.g. using density functional theory is an ideal combination to obtain a fundamental insight into the structure-properties relationship of graphene-derived nanostructures.

Here, I will briefly review the concept of on-surface synthesis as a versatile tool to create nanographene materials previously inaccessible via wet chemistry routes due to insolubility or reactivity of the final structures. Next, I will discuss the concept of localized topological states in GNRs, which can occur at their ends, hetero-junctions or edge extensions. By creating well defined periodic sequences of these topological electronic modes, one-dimensional electronic bands can be created, which are described by the Su-Schrieffer-Heeger (SSH) Hamiltonian representing the dimerized atomic chain [1]. A strategy to realize small band gap 1D GNR and polymers using the concept of a topological phase transition in a GNR structure family as well as their experimental realization and characterization by scanning tunneling microscopy and spectroscopy of such chains will be presented [2,3]. Finally, I will discuss the magnetic properties of localized π -electron states [4,5].

References

- [1] O. Gröning et al., Nature 560, 209 (2018).
- [2] Q. Sun et al., Adv. Matter, 32, 1906054 (2020)
- [3] B. Cirera et al., Nat. Nanotechnol. 15, 437 (2020)
- [4] Q. Sun et al., Nano Letters, 20, 6429 (2020)
- [5] S. Mishra et al., Nat. Nanotechnol. 15, 22 (2020)

Molecular Magnetism on Surfaces

M. Gruber¹

¹*Faculty of Physics, University of Duisburg-Essen, Duisburg, Germany*

Magnetic molecules on surfaces have attracted considerable interest, in particular, in view of potential (quantum) technological applications. The ligands around the magnetic center essentially determine the magnetic properties of the metal center, and to a certain extent, protect them against the environment. Magnetic molecules are thus also seen as fascinating prototypical objects to address fundamental physics questions.

Spin-crossover complexes are a class of molecular systems exhibiting two stable configurations of the ligands and two corresponding spin states of the metal center. We will present the reversible spin-state switching of such complexes on Ag(111) via injection of tunneling electrons [1]. We will show that the molecular stacking is particularly important and can prohibit the switching. The ligand field is often believed to quench the orbital moment of the metal center. However, the remaining orbital moment depends on the details of the *d*-orbital splitting. For a complex composed of two Co ions deposited on Ag(111), we show that the orbital moment is of the same order of magnitude than the spin moment, despite a ligand field on the order of an electronvolt [2]. The interactions of neighboring molecules may lead to small change of the molecule's magnetic moment, but these small changes are often difficult to monitor. We used the interaction of the molecule's magnetic moment with Cooper pairs of a superconducting substrate (so-called Yu-Shiba-Rusinov resonances) to monitor such tiny changes in the magnetic moment [3].

References

- [1] S. Johannsen, S. Ossinger, T. Markussen, F. Tucek, M. Gruber, R. Berndt, *ACS Nano* 15, 11770 (2021)
- [2] C. Li, R. Robles, N. Lorente, S. Mahatha, S. Rohlf, K. Rosnagel, A. Barla, B. Sorokin, S. Rusponi, P. Ohresser, S. Realista, P. Martinho, T. Japser-Toennies, A. Weismann, R. Berndt, M. Gruber, *ACS Nano* 17, 10608 (2023)
- [3] J. Homberg, A. Weismann, R. Berndt, M. Gruber, *ACS Nano* 14, 17387 (2020)

SPIN ENGINEERING IN ARTIFICIAL ATOM-MOLECULE HYBRIDS

**Wantong Huang¹, Máté Stark¹, Paul Greule¹, Daria Sostina²,
Constantin Weeber¹, Jose Gálvez³, Christoph Sürgers¹, Christoph
Wolf³, Wolfgang Wernsdorfer^{1,2}, and Philip Willke¹**

¹*Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany*

²*Institute of Quantum Materials and Technologies (IQMT), Karlsruhe Institute of
Technology, Karlsruhe, Germany*

³*Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic
of Korea*

The combination of electron spin resonance (ESR) and scanning tunneling microscopy (STM) allows to address single spins on surfaces with unprecedented energy resolution and to control them coherently [1]. For this technique, both individual atoms as well as magnetic molecules [2,3], consisting of one central spin surrounded by ligands, are promising building blocks and offer a route for nanoscale spintronics and future quantum devices. Here, we explore the magnetic properties and spin dynamics of artificially built hybrids of atomic and molecular spins. We construct atomic dimers consisting of iron phthalocyanine (FePc) molecules and Fe atoms adsorbed on magnesium oxide by STM tip manipulation. We investigate their emergent magnetic properties by both scanning tunneling spectroscopy and ESR-STM. We believe, that such magnetic prototype structures provide a versatile platform to study complex magnetic interactions and atomic-scale spin dynamics.

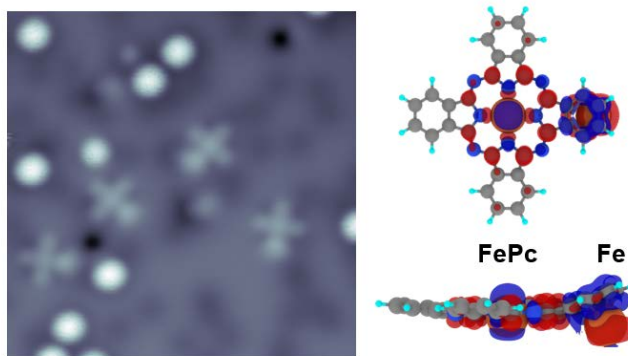


Figure 1: Left: STM topography image (9 nm x 9 nm) of four artificially built dimers of Fe and FePc using the STM tip. Right: DFT calculations of the geometry arrangement in the dimer.

References

- [1] Chen, Y., Bae, Y., A.J., et al., *Adv. Mat.*, 2107534 (2022)
- [2] Zhang, X., Wolf, et al., *Nat. Chem.*, 14(1), 59-65 (2022)
- [3] Willke, P., Bilgeri, T., et al., *ACS nano*, 15(11), 17859-17965 (2021)

A Universal Model for STM-Induced Luminescence

Song Jiang,¹ Tomáš Neuman,² Rémi Bretel,² Alex Boeglin,¹

Fabrice Scheurer,¹ Eric Le Moal,² and Guillaume Schull¹

¹ *Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France*

² *Institut des Sciences Moléculaires d'Orsay (ISMO), UMR 8214, CNRS, Université Paris-Saclay, 91405 Orsay Cedex, France*

Fluorescence of neutral and charged molecules has been reported with subnanometer resolution in scanning tunneling microscope induced-luminescence (STML) experiments over the last years.[1] Despite these significant developments, the detailed mechanisms of STML of individual molecules remained to be fully understood. In these studies, an ultrathin decoupling layer is introduced between the molecules and the supporting metal sample to suppress the substrate-induced fluorescence quenching. This strategic approach has also been employed for precise control of the charge states of single molecules,[2] which suggests that different charge states should be considered in STML process.

Here, we proposed a “universal” fluorescence model relying on successive carrier tunneling events that is based on a systematic experimental STML study of quinacridone (QA) molecules adsorbed on four-monolayer (4 ML) NaCl/Ag(111).[3] A rare property of this molecule is that neutral and positively charged fluorescence dipoles are oriented at $\approx 65^\circ$ from each other. This feature allows us to unambiguously distinguish the charged and neutral emissions solely from their distinct patterns in STML maps.

Based on a comparison between the spatial, bias voltage, and tunneling current dependences of the charged and neutral emission, and on conductance data, a many-body description of the system is set. It reveals that four different charged states of QA (QA^- , QA^0 , QA^+ , and QA^{2+}), involving three different spin multiplicities, can be populated within a single voltage sweep. This excitation mechanism is backed up by data obtained for QA deposited on NaCl/Au(111) where the ground state of the molecule is a doubly positively charged singlet state (S^{2+}). This model bears a universal character that can be applied to STML experiments dealing with charged and neutral molecules.

References

- [1] X. H. Qiu, et al., *Science* **299**, 542-546 (2003) ; Y. Zhang, et al., *Nature* **531**, 623-627 (2016) ; H. Imada et al., *Nature* **538**, 364-367 (2016) ; B. Doppagne et al., *Science* **361**, 251-255 (2018).
- [2] P. Scheurer, et al., *Phys. Rev. Lett.* **123**, 066001 (2019). K. Kaiser, et al., *Nat. Commun.* **14**, 4988 (2023).
- [3] S. Jiang, et al., *Phys. Rev. Lett.* **130**, 126202 (2023)

Desorption from surfaces: Impact of molecular-level details on desorption kinetics

A. Kühnle¹

¹*Physical Chemistry I, Bielefeld University, Bielefeld, Germany*

Desorption of molecules from surfaces is a ubiquitous process both in nature and technology. Despite its omnipresence and conceptual simplicity, fundamental details can be rather complex. Often first-order kinetics are assumed, implying independent, single-step desorption events. While this might be a good approximation in some instances, it is far from reality for many systems, e.g., when molecules can exchange between first and second layer [1] or when different adsorption sites are present [2]. Moreover, molecules that form ordered layers are subject to intermolecular interactions. In this case, the local molecular arrangement, the interaction range and its dependence on coverage can result in a complex desorption behavior [3]. Based on isothermal atomic force microscopy imaging, the desorption can be followed by collecting a series of images and tracing the coverage. These series reveal not only the desorption rate as a function of time, but also local information that shed light onto the molecular desorption mechanism.

In this talk, fundamental concepts of molecular desorption will be reviewed and selected examples will be presented [1-3] to illustrate the variety of molecular desorption phenomena.

References

- [1] Tobias Dickbreder *et al.*, Phys. Chem. Chem. Phys. **23** 18314 (2021)
- [2] Tobias Dickbreder *et al.*, Phys. Chem. Chem. Phys. **25** 12694 (2023)
- [3] Florian Schneider *et al.*, in preparation

Hamiltonian learning and triplon quantum matter in atomically assembled spin lattices

Jose Lado

Department of Applied Physics, Aalto University

Atomic-scale quantum-magnets provide a versatile platform to explore the emergence of quantum excitations in many-body systems. The engineering of spin models at the atomic scale with scanning tunneling microscopy and the local imaging of excitations with electrically driven spin resonance has risen as a powerful strategy to image spin excitations in finite quantum spin systems. First [1], we show that dynamical spin excitations provide a robust strategy to infer the nature of the underlying Hamiltonian, and to design artificial spin modes. We show that finite-size interference of the dynamical many-body spin excitations of a generalized long-range Heisenberg model allows the underlying spin couplings to be inferred. We show that the spatial distribution of local spin excitations directly correlates with the underlying ground state in the thermodynamic limit. Our results put forward local dynamical excitations in confined quantum spin models as versatile witnesses of the underlying ground state, providing an experimentally robust strategy for Hamiltonian inference in complex real spin models. Second [2], we demonstrate the emergence of triplon excitations in an artificial atomic-scale magnet, exotic many-body modes emerging from propagating singlet-triplet transitions. By engineering a minimal quantum magnet, we demonstrate the emergence of dispersive triplon modes in one- and two-dimensional assemblies probed with scanning tunneling microscopy and spectroscopy. Our results provide the first demonstration of dispersive triplon excitations from a real-space measurement. These results establish the potential of artificial spin models to engineer quantum many body matter, and establish Hamiltonian learning as a powerful tool to design artificial quantum magnets.

[1] Netta Karjalainen, Zina Lippo, Guangze Chen, Rouven Koch, Adolfo O. Fumega, and Jose L. Lado, *Phys. Rev. Applied* 20, 024054 (2023)

[2] Robert Drost, Shawulienu Kezilebieke, Jose L. Lado, and Peter Liljeroth, *Phys. Rev. Lett.* 131, 086701 (2023)

Theory of single-impurity electron spin resonance on surfaces

Nicolás Lorente^{1,2}

¹*Centro de Física de Materiales, CFM/MPC (CSIC-UPV/EHU), Paseo de Manuel de Lardizabal 5, 20018 Donostia-San Sebastián, Spain*

²*Donostia International Physics Center (DIPC), Paseo de Manuel de Lardizabal 4, 20018 Donostia-San Sebastián, Spain*

Recent progress in electron spin resonance with the scanning tunneling microscope (ESR-STM) [1] is greatly advancing the experimental possibilities of manipulating atomic spins by all-electrical means. Two-qubit operations have been made possible using a pulse-mode in the ESR-STM [2], and addressing remote qubits has been rendered possible by creating a new multi-frequency operational mode [3]. Using the STM implies detecting electronic currents that contain information on the qubit operations. The way this information is encoded in the current can be complex and developing a computational tool that permits us to interpret and predict the outcome of experiments in ESR is of uttermost importance.

The first results of such a simulation tool have addressed a different number of spins under an STM current [4-6]. We use a non-equilibrium Green's function approach with Hubbard operators that allows us to write quantum adiabatic Markovian master equations in the presence of an electron current and under the driving of an external electric field. The results are enticing with a pleasant agreement with existing experiments.

References

- [1] S. Baumann et al, *Science* **350**, 417 (2015)
- [2] K. Yang et al, *Science* **366**, 509 (2019)
- [3] S.-H. Phark et al, *ArXiv:2108.09880*
- [4] J. Reina et al, *Phys. Rev. B* **100**, 035411 (2019)
- [5] J. Reina et al, *Phys. Rev. B* **104**, 245435 (2021)
- [6] J. Reina et al, *Phys. Rev. B* **107**, 235404 (2023)

From high-resolution molecular spectroscopy to moiré-tuned spin interactions of individual atoms: MoS₂/Au(111) as a versatile lab bench for STM studies

Christian Lotze¹ and co-authors in refs. [3] to [7]

¹*Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany*

MoS₂ can be grown on Au(111) by CVD in ultraclean single layers [1]. While single layer MoS₂ is a direct band gap semiconductor, MoS₂ adsorbed on a metallic substrate is prone to substantial hybridization with the underlying substrate [2], yet, on Au(111) featuring a band gap of below ~2 eV [3]. Because of the lattice mismatch between MoS₂ and the underlying Au(111), a moiré superlattice of topographic and electronic nature is formed.

Yet, MoS₂ on metallic substrates can act as an effective electronic decoupling layer for molecular adsorbates also because it exhibits a small electron-phonon coupling strength. As such, employing tunnelling spectroscopy allows to resolve spectroscopic features like vibronic excitations with spectral resolution close to the thermal limit for molecular states that lie in its band gap [4,5].

However, for magnetic adsorbates like individual Fe adatoms the interaction with the conduction electrons of the underlying Au substrate varies depending on the position with respect to the MoS₂ moiré superlattice and covers the range from unperturbed, pure inelastic spin-flip excitations to fully Kondo-screened [6].

For Mn adatoms also the interatomic coupling can be tuned by manipulation of the distance between two atoms and we can investigate their properties in the strong and weak exchange coupling regime. Here, we find that at sufficiently close spacings, the Mn atoms exhibit a strong coupling and form a non-magnetic singlet ground state, which is unaffected by Kondo-like exchange coupling but do influence the properties of the excited magnetic triplet state [7].

References

- [1] S. G. Sørensen et al., ACS Nano, **8**, 7, 6788 (2014)
- [2] A. Bruix et al., Phys. Rev. B **93**, 165422 (2016)
- [3] N. Krane et al., Nano Lett. **16**, 5163 (2016)
- [4] N. Krane et al., ACS Nano, **12**, 11, 11698 (2018)
- [5] G. Reecht et al., Phys. Rev. Lett. **124**, 116804 (2020)
- [6] S. Trishin et al., Phys. Rev. Lett. **127**, 236801 (2021)
- [7] S. Trishin et al., Phys. Rev. Lett. **130**, 176201 (2023)

Artificial quantum states in assembled van der Waals heterostructures studied by scanning tunneling microscopy

F. Lüpke

Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany

The assembly of van der Waals (vdW) materials into heterostructures enables the engineering of exotic quantum states by moiré and proximity effects. Scanning tunneling microscopy (STM) has become an important tool to study the properties of such heterostructure, because it gives direct access to the resulting structural and electronic properties. While the possibilities to combine individual vdW layers in Lego-like fashion are virtually infinite, the fabrication of samples with atomically clean surfaces, as required for high-resolution scanning probe studies, was typically limited to air-stable or epitaxially grown samples, which drastically diminished the options. In my talk, I will summarize our developed vdW assembly methods [1], which allow the stacking of air-sensitive vdW materials with atomically clean surfaces and interfaces, and will show results of proximity heterostructures [2,3] and twisted bilayers [4].

References

- [1] K. Jin, *et al.*, arXiv.2306.10305 (2023)
- [2] F. Lüpke *et al.*, *Nature Physics* **16**, 526 (2020)
- [3] J. Martinez-Castro *et al.*, arXiv.2304.08142 (2023)
- [4] F. Lüpke *et al.*, *Nano Letters* **14**, 5674 (2022)

Acknowledgements

The authors acknowledge support by the German Research Foundation's (DFG) Emmy Noether Programme and Priority programme 2244 ('2D Materials – Physics of van der Waals [hetero]structures', project nos. 443416235 and 422707584); the Alexander von Humboldt Foundation; Germany's Excellence Strategy - Cluster of Excellence Matter and Light for Quantum Computing (ML4Q); European Union's Horizon 2020 Research and Innovation Programme under Grant Agreement no 824109 (European Microkelvin Platform); the Helmholtz Nano Facility; APVV-20-0425, VEGA 2/0058/20, Slovak Academy of Sciences project IMPULZ IM-2021-42, COST action CA21144 (SUPERQUMAP) and EU ERDF (European regional development fund) Grant No. VA SR ITMS2014+ 313011W856.

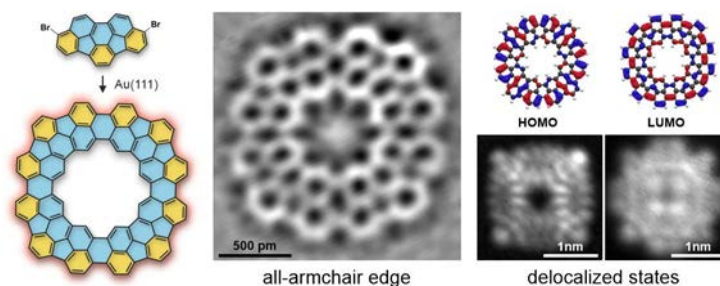
ON-SURFACE SYNTHESIS: A BOTTOM-UP STRATEGY TO ATOMICALLY PRECISE MATERIALS

S. Maier¹

¹Friedrich-Alexander-University Erlangen-Nürnberg

On-surface synthesis via covalent coupling of adsorbed molecules on metal surfaces has attracted significant attention recently due to its potential to fabricate low-dimensional carbon materials with atomic precision. To understand and control the surface-chemistry-driven synthesis, many efforts have been made to design smart precursors, explore novel reaction schemes, and utilize templating effects from the substrate.

My presentation focuses on high-resolution scanning probe microscopy experiments combined with density functional theory to demonstrate recent highlights on the assembly and characterization of physical properties of surface-supported low-dimensional molecular structures. First, the assembly and electronic structure of planar π -extended cycloparaphenylene macrocycles, which represent the first nanographene with an all-armchair edge topology, will be discussed.[1] In the second part, I will outline the host-guest chemistry and the electronic properties of covalent and metal-organic 2D networks that are assembled via debromination reactions on metal surfaces.[2-5]



Planar π -extended cycloparaphenylene synthesized by on-surface [1]

References:

- [1] F. Xiang, et al., Nature Chem., 2022 14, 871–876.
- [2] C. Steiner, et al., Nature Commun., 2017 8, 14765.
- [3] Z. Yang, et al., ACS Nano 2020 14, 12, 16887-16896.
- [4] Z. Yang, et al. Angew. Chem. Int. Ed. 2020, 59, 24, 9549-9555.
- [5] Z. Yang et al. Nanoscale, 2021 13, 9798–9807.

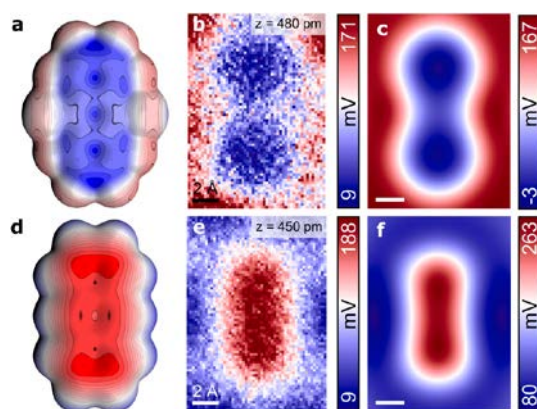
Visualization of π -hole in molecules by means of Kelvin probe force microscopy

Benjamin Mallada, Martin Ondráček, Maximilian Lamanec, Aurelio Gallardo, Alejandro Jiménez-Martín, Bruno de la Torre, Pavel Hobza, Pavel Jelínek

*Regional Centre of Advanced Technologies and Materials,
Czech Advanced Technology and Research Institute (CATRIN), Palacký University
Olomouc, 78371 Olomouc, Czech Republic.*

mallada@fzu.cz

Submolecular charge distribution significantly affects the physical-chemical properties of molecules and their mutual interaction. One example is the presence of a π -electron-deficient cavity in halogen-substituted polyaromatic hydrocarbon compounds, the so-called π -holes, the existence of which was predicted theoretically [1], but the direct experimental observation is still missing. Here we present the resolution of the π -hole on a single molecule using the Kelvin probe force microscopy [2], which supports the theoretical prediction of its existence. In addition, experimental measurements supported by theoretical calculations show the importance of π -holes in the process of adsorption of molecules on solid-state surfaces. This study expands our understanding of the π -hole systems and, at the same time, opens up possibilities for studying the influence of submolecular charge distribution on the chemical properties of molecules and their mutual interaction.



References

- [1] Politzer, P. & Murray, J. S. Electrostatics and Polarization in σ and π - Hole Noncovalent Interactions: An Overview. *ChemPhysChem* vol. **21** 579–588 (2020).
- [2] Mallada, B., Ondráček, M., Lamanec, M. et al. Visualization of π -hole in molecules by means of Kelvin probe force microscopy. *Nat Commun* **14**, 4954 (2023)

Scanning tunneling spectroscopy of 2D materials: Quantum Hall edge states and topological magnetic gaps

Markus Morgenstern (RWTH Aachen University)

The talk will focus on the abilities of low-temperature scanning tunneling microscopy to probe van-der-Waals materials using the examples of graphene or MnSb_2Te_4 . For graphene, I will show how one can avoid the tip-induced quantum dot (TIQD) during the mapping of quantum Hall edge states by carefully choosing gate voltages after examining the properties of the TIQD in detail by using its charging lines and its probed states [1]. This approach opens the door to study the topological edge states in the quantum Hall regime (Fig. 1) and its reconstructions on the relevant nm length scale. MnSb_2Te_4 with a slight excess of Mn (6 %) is a ferromagnetic topological insulator with its Dirac cone close to the Fermi level, where a magnetic gap opens below the Curie temperature $T_C=45$ K [2]. STM reveals the relatively strong magnetic disorder by a spatially varying gap size (Fig. 2) at largely homogeneous gap center, the partial closure of the gap by in-plane magnetic field and spatiotemporal fluctuations of the gap size at elevated temperature [3].

[1] Phys. Rev. B 107, 115426 (2023) [2] Adv. Mater. 33, 2102935 (2021) [3] arXiv:2107:04428

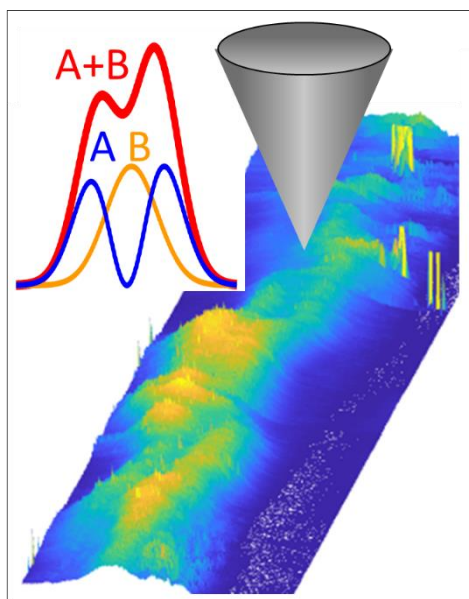


Fig. 1: 3D representation of a probed edge state belonging to Landau level -2 probed at the Fermi level and $B = 7$ T. The inset shows the theoretical cross section of the edge state with its two contributions being sublattice-polarized for each valley.

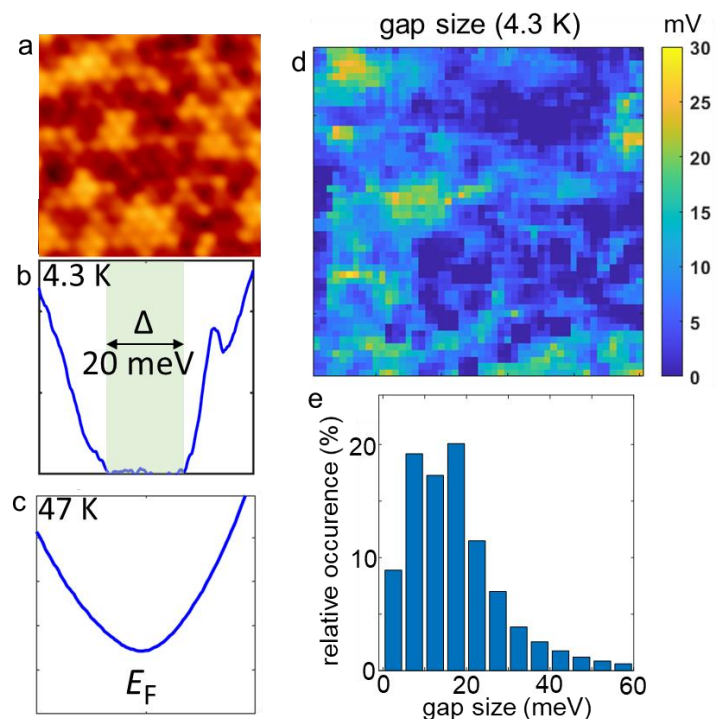


Fig 2: (a) Atomically resolved topography of Mn-rich MnSb_2Te_4 ($5 \times 5 \text{ nm}^2$). (b), (c) Individual dI/dV spectra at the indicated temperatures. (d) Map of the gap size at 4.3 K ($20 \times 20 \text{ nm}^2$) (e) histogram of gap sizes originating from several maps.

Atomic spins on surfaces: from early investigation to coherent control

Sander Otte¹

¹Delft University of Technology, Delft, The Netherlands

A scanning tunneling microscope is capable of measuring just a single quantity: the tunneling current. Yet, when it comes to the study of individual magnetic atoms on a surface, this quantity has proven to carry a surprising amount of valuable information. In this keynote presentation, I will give an overview of two decades of research on spins on surfaces. Starting from the first spin-polarized measurements and incoherent excitations on magnetic atoms, I will discuss a number of techniques that eventually led to the possibility to perform electron spin resonance on individual atoms. Particular focus will be given to recent developments in my group on pump-probe spectroscopy, resulting in the capability to trace the coherent dynamics of coupled spins in real time over the course of tens of nanoseconds. I will end the talk with an outlook of future prospects for this blooming field.

Quantum features of magnetic molecules on surfaces: from “innocent” surfaces to type I superconductors.

Lorenzo Poggini^{1,2}

¹ *Institute for Chemistry of OrganoMetallic Compounds (ICCOM-CNR), 50019 Fiorentino (FI), Italy.*

² *Department of Chemistry “Ugo Schiff” (DICUS), Università degli Studi di Firenze, 50019 Sesto Fiorentino (FI), Italy.*

Single Molecule Magnets and Magnetic Molecules (SMMs and MM respectively) are inorganic molecules having remarkable magnetic properties, such as extremely long magnetization relaxation time and magnetic hysteresis at the single molecule (or ion) level at low temperatures.^[1] Thanks to these properties, they represent suitable building blocks for the advanced nanoscale technologies of spintronics and quantum computing. In this context, novel phenomena attractive phenomena arose from coupling magnetic materials to superconductors.^[2] Here we show a microscopic and spectroscopic study of a sub-monolayer of Fe₄ SMMs deposited on a Pb(111) surface. We reveal that the transition of lead to the superconducting state affects the magnetization of the SMMs that locally switch from a blocked magnetization state to a resonant quantum tunnelling regime.^[3] Moreover, we demonstrated that a single layer of magnetic molecules, Terbium(III) bis-phthalocyaninato (TbPC₂) complexes, deposited on Pb(111) surface is sensitive to the topology of the intermediate state of the superconductor imprinting a local evolution of the magnetization of the TbPC₂ molecules in the monolayer.^[4] Concerning molecular qubits, a noteworthy candidate showing coherence times in the order of tens of μs, is (η⁸-cyclooctatetraene)(η⁵-cyclopentadienyl)titanium, hereafter [CpTi(cot)].^[5] We report a morphological and spectroscopic study of a [CpTi(cot)] monolayer deposited in Ultra High Vacuum on Au(111) and on the superconducting Pb(111). Interestingly, Scanning Tunneling Microscopy measurements evidence that molecules organize in a non-trivial pattern on both surfaces^[6,7] generating a new antibonding orbital that crosses Fermi energy, which is the origin of Shiba coupling on the superconductors.^[7] These findings open intriguing perspectives to employ Magnetic Molecules for sensing local magnetic field variations in molecular/superconductor hybrid devices.

References

- [1] D. Gatteschi, R. Sessoli, *Molecular Nanomagnets*, Oxford University Press, 2006.
- [2] J. Linder, J. W. A. Robinson, *Nat. Phys.* 2015, *11*, 307–315.
- [3] G. Serrano, L. Poggini, M. Briganti, et al., *Nat. Mater.* **2020**, *19*, 546–551.
- [3] E. Moreno Pineda, T. Komeda, et al. *Dalton Trans.* 2016, *45*, 18417–18433.
- [4] G. Serrano, L. Poggini, et al., *Nat. Commun.* 2022, *13*, 3838.
- [5] L. C. Camargo, M. Briganti, et al., *Angew. Chemie* 2021, *133*, 2620–2625.
- [9] M. Briganti, G. Serrano, L. Poggini, et al., *Nano Lett.* 2022, *22*, 8626–8632.
- [10] L. Arnhold, M. Briganti, N. Betz, G. Serrano, L. Poggini, et al, Article in preparation.

Measuring Chemical Reactivity at the Atomic Scale

Alexandre Riss

Physics Department E20, TUM School of Natural Sciences, Technical University of Munich, D-85748 Garching, Germany

Email: a.riss@tum.de

Despite significant advances in theoretical modeling, understanding the chemical reactivity of organic molecules and deriving quantitative predictions from structural models remains a challenge. In many cases chemical intuition and heuristics are relied upon to assess reactivity. In this presentation, I will demonstrate how high-resolution atomic force microscopy (AFM) can be employed to quantify chemical reactivity and interaction potentials, providing accurate insights into the behavior of organic molecules.

In particular, we investigated the impact of aromaticity on chemical reactivity using porphyrins as a case study (Fig. 1 a,b) [1]. Moreover, we show interaction potentials between individual reactive sites and specific probes attached to AFM tips can be directly measured (Fig. 1c), giving a more detailed understanding of the atomic-scale forces that govern chemical reactions.

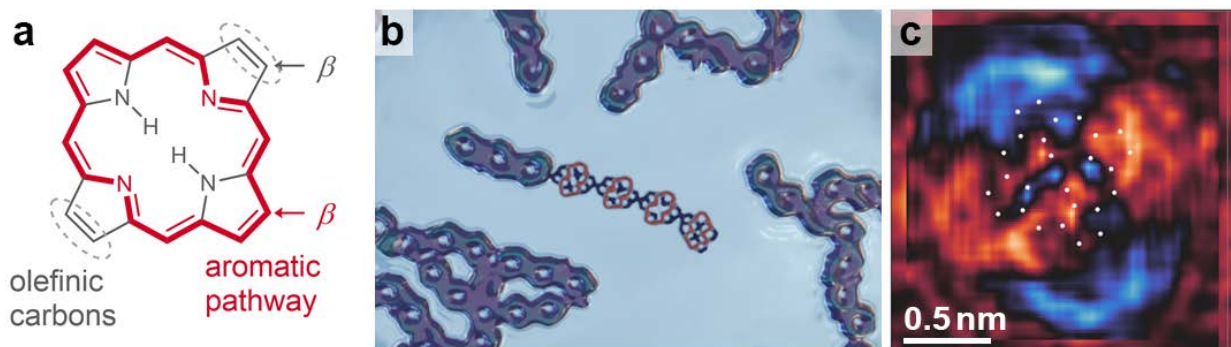


Fig. 1: Effect of aromaticity on the chemical reactivity of surface-supported porphyrins. **(a)** Aromatic pathway in a free-base porphyrin, giving rise to two chemically different types of β -carbons at the “corners” of the macrocycle. **(b)** 3D rendering of STM and AFM measurements of the products of cyclization and coupling reactions of alkyne-substituted porphyrins. These experiments reveal a substantially lower reactivity of the β -carbon atoms within the aromatic pathway. **(c)** Interaction forces between a free-base porphyrin (atomic positions overlaid in white color) and a functionalized AFM tip, showing a twofold symmetry.

References

[1] N. Cao et al. *Nat. Chem.* 2023: <https://doi.org/10.1038/s41557-023-01327-6>

Light-matter interaction and photochemistry probed with sub-molecular precision

A. Rosławska^{1,2}

¹*Max Planck Institute for Solid State Research, Stuttgart, Germany*

²*Université de Strasbourg, CNRS, IPCMS, UMR 7504, Strasbourg, France*

Light-matter interaction plays a crucial role in the quantum properties of light emission from single molecules, and in electron-to-photon and photon-to-electron energy conversion. Recent works have shown that both the tunneling current of an STM and incoming laser radiation can be used to excite the intrinsic luminescence of individual molecules enabling studying light-matter interaction with unprecedented resolution. In the talk, I will first discuss how STM-induced luminescence allows us to map the optical properties of single molecules with nearly atomic precision [1]. With this approach, we are also able to mimic structures employed by photosynthetic systems and study sub-nm details of the energy transfer process using molecules as ancillary, passive or blocking elements to promote and direct resonant energy transfer between distant donor and acceptor units [2]. Furthermore, using a tunable light source focused on the STM tip we drive and control the rate of a free-base phthalocyanine phototautomerization with sub-molecular precision, providing a path to pilot the intrinsic reactivity of the molecule with an external controllable stimulus. We probe the excited states of the molecule, which drive the tautomerization, by recording tip-enhanced photoluminescence (TEPL) spectra and maps where varying patterns for non-resonant and resonant excitation conditions are observed [3]. Such control over optical properties at the molecular level is possible only using this hyper-resolution optical approach, which does not suffer from ensemble averaging.

References

- [1] A. Rosławska *et al*, *Phys Rev. X*, **12**, 011012 (2022).
- [2] S. Cao *et al.*, *Nat. Chem.*, **13**, 766-770 (2021).
- [3] A. Rosławska *et al*, *arXiv:2305.1315* (2023).

Cross-Correlation Microscopy of Atomic Defects in in Transition Metal Dichalcogenides

L. Huberich¹, J. Allerbeck¹, L. Bobzien¹, E. Ammerman¹, F. Xiang¹, O. Gröning¹, Y.-C. Lin², R. Hennig³, D. Qiu⁴, J. Robinson² and B. Schuler¹

¹*Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland*

²*Department of Materials Science and Engineering, The Pennsylvania State University, University Park, USA*

³*Department of Materials Science and Engineering, University of Florida, USA*

⁴*Department of Mechanical Engineering and Materials Science, Yale University, USA*

Two-dimensional (2D) semiconductors provide an exciting platform to engineer atomic quantum states in a robust, yet tunable solid-state system. In this talk, I will present our efforts to unravel the interesting physics behind single dopant atoms in transition metal dichalcogenide (TMD) monolayers by means of high-resolution scanning probe microscopy [1-8].

Our recent research on transition metal doped TMDs such as (n-type) Re-doped MoS₂ and (p-type) V-doped WSe₂ reveals the significance of the charge state in the spectroscopic signature of these defects. By substrate chemical gating, we can stabilize three charge states of Re_{M₀}, where two of the charge states exhibit symmetry broken electronic orbitals and a distorted atomic configuration that we assign to a pseudo Jahn-Teller effect [9]. Negatively charged V dopants and dopant pairs in WSe₂ exhibit a series of occupied p-type defect states above the valence band edge, accompanied by an intriguing electronic fine-structure that we attribute to many-body electron interactions [10].

I will also introduce our efforts to probe excitation dynamics on the ultrafast time scales using a newly developed THz-pulse-driven STM [11].

References

- [1] B. Schuler et al., Phys. Rev. Lett. 123, 076801 (2019)
- [2] B. Schuler et al., ACS Nano 12, 10520 (2019)
- [3] S. Barja et al., Nat. Commun. 10, 3382 (2019)
- [4] K. Cochrane et al., 2D Mater. 7, 031003 (2020)
- [5] M. Aghajanian et al., Phys. Rev. B 101 081201(R) (2020)
- [6] B. Schuler et al., Sci. Adv. 6, eabb5988 (2020)
- [7] K. Cochrane et al., Nat. Commun. 12, 7287 (2021)
- [8] E. Mitterreiter et al., Nat. Commun. 12, 3822 (2021)
- [9] F. Xiang*, L. Huberich* et al. Arxiv: 2308.0220 (in review)
- [10] S. Stolz et al. ACS Nano (in press)
- [11] J. Allerbeck et al. (in review)

Interfacing magnetism and superconductivity: visualizing interactions from 0d to 2d

Paolo Sessi

Max Planck Institute for Microstructure Physics, 06120, Halle (Saale), Germany

Magnetism and superconductivity have been for very long time considered to be two mutually exclusive phases of matter. However, there is constantly increasing evidence that, under the right conditions, these competing phases can coexist. I will discuss a series of experiments where, using a bottom up approach, the interactions between local magnetic perturbations coupled to a superconducting condensate are visualized at the atomic scale. Starting from the 0D case, I will show how it is possible to systematically tune the energy of the so called Yu-Shiba-Rusinov states induced inside the superconducting gap by changing the 3d transition metal element, elucidating trends which are directly linked to the strength of magnetic exchange scattering perturbing the Cooper pairs. Subsequently, I will show how the use of anisotropic surfaces allows to tune the interaction between magnetic dimers coupled through the superconducting substrate, giving rise to a large variety of different configurations. Based on these findings, I will demonstrate how it is possible to engineer different Yu-Shiba-Rusinov bands in 1D spin chains and discuss their relevance within the context of topological superconductivity. Finally, I will present recent experiments on artificial 2D lattices showing how the choice of different lattice symmetries impacts onto the superconducting state and how the design of distinct terminations offers a new control knob to verify the bulk-boundary correspondence characterizing topological states.

Imaging and Tracking Single Polarons by STM/AFM

M. Setvin¹

¹*Charles University, Prague, Czech Republic*

Noncontact atomic force microscopy (nc-AFM) has recently proven to be a powerful tool for manipulating and imaging single electrons at surfaces. Single electrons could be localized at trapping sites such as Au atoms on thin NaCl films [1], small molecules on oxide surfaces [2,3], or larger organic molecules on thicker NaCl films [4].

This talk will focus on the possibilities of investigating polarons in materials: Polarons are electrons or holes that self-localize in ionic lattices due to electron-phonon interaction [5]. Such charge carriers can move to adjacent lattice positions, provided they are given certain activation energy. Polarons play a key role in many applications and materials properties, such as electrical conductivity, optical properties, catalysis and photocatalysis, and they stand behind exotic properties such as colossal magnetoresistance or high-temperature superconductivity. The successful imaging of polarons in hematite Fe_2O_3 and SrTiO_3 will be shown, and the new possibilities offered by this technique will be discussed.

References

- [1] L. Gross et al., *Science* **324**, 1428 (2009)
- [2] M. Setvin et al., *PNAS* **114**, E2556 (2017)
- [3] Z. Wang et al., *Science Advances* **8**, eabq1433 (2022)
- [4] S. Fatayer et al., *Science* **365**, 142 (2019)
- [5] C. Franchini et al., *Nature Reviews Materials* **6**, 560 (2021)

Quantum Sensing with Single Spin Sensitivity

A. Singha¹

¹Max Planck Institute for Solid State Research, Stuttgart, Germany

Controlling individual atoms and molecules at their native spatio-temporal limit has an indispensable appeal that has driven fundamental research for decades, leading to technological leaps ranging from nanoelectronics to information processing. Recently, a new avenue has emerged for nanoscale quantum bits made of individual surface-supported lanthanide atoms^[1] and molecules. The large magnetic moments, strong intra- as well as interatomic electron correlations, and long-term quantum coherence in these systems altogether offer a myriad of new exciting possibilities across molecular magnetism, quantum computing, as well as quantum sensing research.

My effort is devoted to manoeuvre quantum properties of such smallest building blocks of matter, especially by probing them in the least invasive manner. In this regard, the recent focus in my group involves the development of advanced scanning probe NV magnetometry, particularly aiming for non-invasive sensing and coherent control of such systems^[2]. This mandates extending the current capabilities of scanning NV-probe for operations under UHV and low temperatures, which we have recently achieved via controlled surface modifications of diamond^[3]. In my talk, I will elaborate on this including our recent work on readout and control of isolated single molecule qubit candidates (N@C₆₀) using NV-magnetometry approach^[2].

References

- [1] A. Singha*, P. Willke*, T. Bilgeri*, X. Zhang, H. Brune, F. Donati, A. J. Heinrich, T. Choi, *Nat Commun.* **12**, 4179 (2021).
- [2] D. Pinto, D. Paone, B. Kern, T. Dierker, R. Wieczorek, A. Singha, D. Dasari, A. Finkler, W. Harneit, J. Wrachtrup, K. Kern, *Nat Commun.* **11**, 6405 (2020).
- [3] J. N. Neethirajan, T. Hache, D. Paone, D. Pinto, A. Denisenko, R. Stöhr, P. Udverhelyi, A. Pershin, A. Gali, J. Wrachtrup, K. Kern, A. Singha, *Nano Lett.* **23**, 2563-2569 (2023)

Unraveling Orbital Magnetism Contributions to Landau Levels in Moiré Quantum Matter

Joseph A. Stroscio^{1*}

¹*Physical Measurement Laboratory, National Institute of Standards and Technology,
Gaithersburg, MD, 20899, USA*

*E-mail: joseph.stroscio@nist.gov

Flat and narrow band physics in moiré quantum matter (MQM) has proven to be extremely rich with new emergent quantum phases which can be tuned with applied electric and magnetic fields. The topological properties of the eigenstates of the moiré Hamiltonian are critical for establishing the quantum phase of the system. For almost a century, magnetic oscillations have been a powerful “quantum ruler” for measuring Fermi surface topology. In this talk, we use Landau level spectroscopy to unravel the energy-resolved valley-contrasting orbital magnetism and large orbital magnetic susceptibility that contribute to the energies of Landau levels of twisted double bilayer graphene. These orbital magnetism effects lead to significant deviations from the standard Onsager relation, which manifests as a breakdown in scaling of Landau level orbits. These substantial magnetic responses emerge from the nontrivial quantum geometry of the electronic structure and the large length scale of the moiré lattice potential. We show that this breakdown of the original Onsager relation is unique in MQM due to the typical superlattice length scales in these systems. Going beyond traditional measurements, STM-based Landau level spectroscopy offers a complete “quantum ruler” resolving the full energy dependence of orbital magnetic properties in moiré quantum matter.

Abstracts of Posters

(in alphabetical order)

Single atom co-catalyst dispersion on perovskite surfaces by polarity compensation

A. Alexander¹, D. Wrana^{1,2}, P. K. Samal¹, J. Škvára¹, L. Albons¹, J. Redondo¹, L. Fusek¹, I. Piš², V. Johánek¹, J. Mysliveček¹, M. Setvin¹

¹*Department of Surface and Plasma Science, Charles University, Prague, Czech Republic*

²*Marian Smoluchowski Institute of Physics, Jagiellonian University, Krakow, Poland*

³*IOM-CNR, Istituto Officina dei Materiali, AREA Science Park Basovizza, Trieste, 34149, Italy*

aji.alexander@mff.cuni.cz

Perovskite surfaces have been attracting attention in the catalysis community due to their promising chemical properties, high ability to separate electron-hole pairs for light harvesting, and the presence of ferroelectricity in many phases [1]. While perovskite crystals possess a unique set of interesting bulk properties, their surfaces are much less understood; the main open questions are their structural stability and associated chemical reactivity and catalytic selectivity.

This poster focuses on the atomic structure of doped $KTaO_3$ – (001) and $SrTiO_3$ – (001) perovskite surfaces and the potential to modify their catalytic activity by the presence of extrinsic metals, with the main focus on cobalt. Techniques such as STM/AFM along with XPS are implemented to show the thermal stability and the tendency of the guest atoms to disperse in the form of single adatoms on the polarity uncompensated $KTaO_3$ -(001) [2] and $SrTiO_3$ -(001) surfaces. The interaction of cobalt with perovskite surfaces under various reducing and oxidizing conditions, and at different temperatures will be highlighted. Our findings help to explain how the occurrence of various phases of supported cobalt (metallic, oxide, and hydroxide) depend on the environment, allowing for control of its catalytic capabilities. One of the most important achievements presented is the ability to control the metal cluster size down to the scale of single atoms, which is one of the biggest challenges in heterogeneous catalysis nowadays.

[1] Cohen, Ronald E. *Nature* 358.6382 (1992)

[2] M. Setvin, M. Reticcioli, F. Poelzleitner et al., *Science* 359, 572 (2018)

The work was supported by projects GAUK 10/252122, GACR 20-21727X and GAUK Primus/20/SCI/009.

Efficient and continuous carrier-envelope phase control for terahertz lightwave-driven scanning tunneling microscopy

J. Allerbeck¹, J. Kuttruff², L. Bobzien¹, L. Huberich¹, M. Tsarev²,
and B. Schuler¹

¹ *nanotech@surfaces, Empa, Swiss Federal Laboratories for Material Science and Technology, Überlandstrasse 129, 8600 Dübendorf Switzerland*

² *University of Konstanz, Universitätsstrasse 10, 78464 Konstanz, Germany*
Email: jonas.allerbeck@empa.ch

THz lightwave-driven scanning tunneling microscopy (THz-STM) has opened up the possibility to study ultrafast dynamics with atomic precision [1]. One of the outstanding challenges in the field is to precisely control the electro-magnetic waveform to selectively excite electronic or vibrational states in low-dimensional material systems, such as molecules or solid-state defects. Here I present a versatile THz-STM toolbox, operating at up to 41 MHz repetition rate and using an efficient method to generate and precisely control the transient waveform of single-cycle THz pulses.

The setup achieves peak THz voltages at the tunneling junction ranging from 1 V at 41 MHz to few-10 V at 1 MHz, where multi-MHz repetition rates enable state-selective tunneling below 0.01 electrons per THz pulse. We benchmark the THz amplitude and waveform using electro-optic sampling, near-field photoemission sampling inside the STM, and precise methods relying on lightwave-driven tunneling. Independent THz lines for pump and probe pulses enable high flexibility for amplitude and phase control, where continuous phase modulation between unipolar and bipolar THz waveforms is established by frustrated internal reflection in right-angle polymer prisms achieving >60% field transmissivity [2].

Preliminary measurements on transition metal dichalcogenide monolayers demonstrate precise calibration of the THz waveform at the tunneling junction, paving the way towards the investigation of charge lifetime in atomic quantum defects [3] with picosecond time resolution.

References

- [1] T. Cocker et al., *Nature* **539**, 263 (2016)
- [2] J. Allerbeck et al., *ACS Photonics* *accepted manuscript* (2023)
- [3] J. Robinson and B. Schuler, *APL* **119**, 140501 (2021)

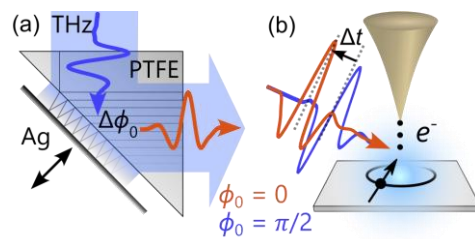


Fig 1: (a) A right angle polymer prism (PTFE) with a metallic mirror modulates the carrier-envelope phase ϕ_0 of single-cycle THz pulses, enabling state-selective tunneling with ultrafast THz fields. (b) Perspective implementation of THz scanning tunneling microscopy with time delayed pulses.

Intricacies in 1T-TaSe₂ Islands with Deposited Cobalt Atoms, A Spinon Kondo Effect?

B. G. Arslan¹, Z. Wang¹, V. Vano² and P. Liljeroth¹

¹*Aalto University, Department of Applied Physics, Espoo, Finland*

²*Princeton University, Department of Physics, New Jersey, USA*

1T-TaSe₂ is a Mott insulator exhibiting a charge density wave (CDW) transition at low temperatures, with a distinctive David's star reconstruction with a 13-Ta-atom unit cell. Scanning tunneling microscopy (STM) reveals upper and lower Hubbard bands (UHB and LHB) on pristine 1T-TaSe₂ islands, indicative of its Mott insulating nature. However, depositing cobalt atoms on 1T-TaSe₂ introduces two additional peaks near the UHB and LHB, interpreted as the spinon Kondo effect [1].

After cobalt deposition, our STM imaging of monolayer 1T-TaSe₂ islands on HOPG reveals cobalt atoms occupying CDW centers, supported by STM topography and dI/dV spectra. The spectra exhibit prominent two additional peaks. Applying an 11 T external magnetic field, expected to impact the spinon band structure, surprisingly results in no changes in the peak energies or line shapes. This suggests that these peaks may have origins other than the spinon Kondo effect.

In summary, we achieved defect-free monolayer growth of 1T-TaSe₂ on HOPG. Our STM investigations verified the presence of the expected additional peaks linked to cobalt deposition. However, external magnetic field experiments did not change these peaks, hinting at alternative mechanisms beyond the spinon Kondo effect. Ongoing research will explore this phenomenon at lower temperatures and with higher energy resolutions.

References

- [1] Y. Chen, et. al. Nat. Phys. 18, 1335–1340 (2022)

Near-Field Amplitude Calibration for THz-STM of Atomic Defects in 2D Semiconductors

L. Bobzien^{1,2}, J. Allerbeck², S. E. Ammermann², B. Schuler²

¹*Institute for Quantum Electronics, Department of Physics, ETH Zurich, 8093 Zurich, Switzerland*

²*nanotech@surfaces Laboratory, Empa, Swiss Federal Laboratories for Material Science and Technology, Überlandstrasse 129, 8600 Dübendorf Switzerland*

The fundamental understanding of quantum dynamics in low-dimensional materials requires simultaneous ultrafast temporal and atomic spatial resolution. Ultrafast scanning tunneling microscopy has emerged as an excellent tool to overcome this challenge. Here, single-cycle THz pulses act as a transient bias, driving temporally confined tunneling currents in a STM. This THz lightwave-driven STM (THz-STM) combines the high spatial resolution and electronic contrast provided by the STM with picosecond temporal resolution of the transient THz field¹. With this system, we thrive to explore 2D semiconductors that attract considerable research interest due to quantum confinement and spin-valley coupling.

A prototypical family of 2D semiconductors are 2D transition metal dichalcogenides (TMDs), where point defects, such as vacancies or impurities, are a perfect model systems to investigate strongly correlated dynamics in low dimensions and offer a rich playground for understanding dynamics in the atomic regime². To investigate point defects in TMDs, our THz-STM combines multi-MHz repetition rates with efficient THz generation for high sensitivity³. I will show recent measurements of the THz rectified charge as a function of bias and THz field amplitude on 2D TMDs. This allows to map the complex energy landscape of localized states with a resolution of down to 0.005 electrons per pulse enabling precise near-field amplitude calibration. Preliminary THz-STM and THz-STS measurements of a sulfur vacancy in MoS₂ demonstrate state-selective tunneling into distinct electronic orbitals of the quantum defect.

The precise control of THz amplitude, polarity, carrier-envelope phase along with appropriate calibration in the optical near field is an enabler for state-selective excitation of local dynamics. This will facilitate the study of charge lifetimes, vibronic coupling and localized quasiparticles in 2D semiconductors at their natural time and length scales.

References

- [1] Cocker, T. *et al.*, Nature **539**, 263–267 (2016).
- [2] Robinson, J. A. & Schuler, B., Applied Physics Letters **119**, 140501 (2021)
- [3] Allerbeck, J. *et al.*, *accepted manuscript*, ACS Photonics (2023)

Electronic Structure of Quasi-Freestanding WS₂/MoS₂ Heterostructures

B. Pielić,¹ D. Novko,¹ I. Šrut Rakić,¹ J. Cai,² M. Petrović,¹
A. Bremerich,² R. Ohmann,² N. Vujičić,¹ M. Basletić,³
M. Kralj,¹ C. Busse²

¹*Institute of Physics, Zagreb, Croatia*

²*Universität Siegen, Siegen, Germany*

³*University of Zagreb, Zagreb, Croatia*

By means of scanning tunneling spectroscopy (STS), we study the electronic structure of monolayer MoS₂, WS₂, and WS₂/MoS₂ vertical heterostructure. Moreover, we investigate a band bending in the vicinity of narrow one-dimensional (1D) interface of the WS₂ – MoS₂ lateral heterostructure. We confirm type-II band alignment for the WS₂ – MoS₂ lateral heterostructure and determine corresponding depletion regions, charge densities, and the electric field at the interface [1].

Epitaxial growth of these lateral WS₂ – MoS₂ and vertical WS₂/MoS₂ heterostructures on graphene under ultra-high vacuum (UHV) conditions enables this in-situ characterization with direct spectroscopic insights. Density functional theory (DFT) is used for the calculation of the band structures, as well as for the density of states (DOS) maps at the interfaces.

Quasi-freestanding heterostructures with sharp interfaces, large built-in electric fields, and narrow depletion region widths are proper candidates for future designing of electronic and optoelectronic devices.

References

- [1] B. Pielić et al., ACS Applied Materials & Interfaces **13**, 50552 (2021)

Electric-field-driven Spin Resonance by an Exchange-coupled Single-atom Magnet

Hong Thi Bui^{1,2}, Alejandro Ferrón³, Joaquin Fernández-Rossier⁴, Jose Reina-Gálvez^{1,2}, Christoph Wolf^{1,2}, Andreas J. Heinrich^{1,2*}, Christopher P. Lutz^{5*}, Soo-hyon Phark^{1,2*}

¹*Center for Quantum Nanoscience, Institute for Basic Science, Seoul 03760, Korea*

²*Department of Physics, Ewha Womans University, Seoul 03760, Korea*

³*CONICET-UNNE and Facultad de Ciencias Exactas, Universidad Nacional del Nordeste, Argentina*

⁴*International Iberian Nanotechnology Laboratory, 4715-330 Braga, Portugal*

⁵*IBM Research Division, Almaden Research Center, San Jose, CA 95120, USA*

Coherent control of individual atomic and molecular spins on surfaces has recently been demonstrated by using electron spin resonance (ESR) in a scanning tunneling microscope (STM). Here we present a combined experimental and modeling study of the ESR of a single hydrogenated Ti atom that is exchange-coupled to a Fe adatom positioned in 0.6-0.8 nm away by means of atom manipulation. Continuous wave and pulsed ESR of the Ti spin show a Rabi rate with two contributions, one from the tip and the other from the Fe, whose spin interactions with Ti are modulated by the radio-frequency electric field. The Fe contribution is comparable to the tip, as revealed from its dominance when the tip is retracted, and tunable using a vector magnetic field. Our new ESR scheme allows on-surface individual spins to be addressed and coherently controlled without the need for magnetic interaction with a tip. This study establishes a feasible implementation of spin-based multi-qubit systems on surfaces.

References

- [1] Soo-Hyon Phark, Adv. Sci. 10, 2302033 (2023)

Coherent Spin Dynamics Between an Electron and Nucleus Within a Single Atom

Lukas M. Veldman¹, Evert W. Stolte¹, Mark P. Canavan¹, Rik Broekhoven¹, Philip Willke², Laëtitia Farinacci¹, Sander Otte¹

¹*Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands.*

²*Physikalisches Institut, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany.*

The combination of electron spin resonance (ESR) and scanning tunneling microscopy (STM) provides a mechanism for studying the interaction between electron and nuclear spins within individual atoms on a surface. Recently, signatures of the hyperfine interaction in TiH on MgO have been observed via splitting of the electron spin transition [1], and has additionally been shown to be highly anisotropic [2]. In this work, we resolve the coherent dynamics originating from the hyperfine interaction between the nuclear and electron spin in a single TiH atom. We tune the local magnetic field at the location of the TiH atom [3] such that the electron and nuclear spins hybridize. Using DC voltage pulses [4] to spin pump the electron and nuclear spin [5], we measure the subsequent coherent free evolution of the coupled electron-nuclear spin system.

References

- [1] Willke et al. Science 362 (2018)
- [2] Farinacci et al. Nano Letters (2022)
- [3] Yang et al. Phys Rev Lett 122 (2019)
- [4] Veldman et al. Science 372 (2021)
- [5] Yang et al. Nature Nanotechnology 13 (2018)

Shot-noise measurements of single-atom junctions using a scanning tunneling microscope

Verena Caspari, Idan Tamir, Daniela Rolf, Christian Lotze, and Katharina J. Franke

Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany

Current passing through small constrictions fluctuates due to the discreteness of charge. Measuring this so-called shot noise in atomic-scale superconducting junctions can provide valuable information, from the quanta of charge transferred in the tunneling process to the correlations of the involved tunneling channels. Here, we use a scanning tunneling microscope equipped with a high-frequency, low-temperature amplifier to measure simultaneously the noise characteristics and the differential conductance of two types of Pb-Pb-junctions having different geometries. The first type is created by deposition and approaching single Pb adatoms, while the second type is established by a break-junction technique. We observe a correlation between the noise pattern and the strength of the Josephson current, independent on the method of junction preparation.

Visualizing the Localized Electrons of a Kagome Flat Band

Caiyun Chen,^{1,2} Jiangchang Zheng,^{1,2} Ruopeng Yu,^{1,2} Soumya Sankar,¹ Hoi Chun Po,¹ Kam Tuen Law,¹ and Berthold Jaeck^{1,*}

¹*Department of Physics, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong SAR*

²*These authors contributed equally.*

Destructive interference between electron wavefunctions on the two-dimensional kagome lattice induces an electronic flat band, which could host a variety of interesting quantum states. Key to realize these proposals is to demonstrate the real space localization of kagome flat band electrons. The extent to which the complex structure of realistic materials counteracts the localizing effect of destructive interference is hitherto unknown. We used scanning tunneling microscopy (STM) to visualize the non-trivial Wannier states of a kagome flat band at the surface of CoSn, a Kagome metal. We find that the local density of states associated with the flat bands of CoSn is localized at the kagome lattice center, consistent with theoretical predictions. Our results show that these states exhibit an extremely small localization length of two to three angstroms concomitant with a strongly renormalized quasiparticle velocity $v \approx 1 \times 10^4$ m/s, comparable to that of moire superlattices. Our findings provide fundamental insight into the electronic properties of kagome metals and present a key step for future research on emergent many-body states in these systems.

References

[1] Caiyun CHEN, arXiv:2308.08976

Nanoscale characterization and light emission aspects of defects in h-BN

F. J. R. Costa^{a,b}, L. F. Zagonel^a, S. Novikov^c, P. H. Beton^c, G. Cassabois^d, A. Rosławska^b, K. Kuhnke^b & K. Kern^{b,e}

^a Gleb Wataghin Institute of Physics - University of Campinas – UNICAMP, Campinas 13083-859, Brazil

^b Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, Stuttgart 70569, Germany

^c School of Physics and Astronomy, University of Nottingham, Nottingham NG7 2RD, United Kingdom

^d Laboratoire Charles Coulomb, UMR5221 CNRS-Université de Montpellier, 34095 Montpellier, France

^e Ecole Polytechnique Fédérale de Lausanne, Lausanne 1015, Switzerland

E-mail: fabiojrc@ifi.unicamp.br

Hexagonal boron nitride (h-BN), akin to graphene in structure, is a wide band-gap layered material, which exhibits captivating optical properties attributed to its lattice defects. These can act as single-photon sources, capable of emitting just one photon per excitation cycle, which is an essential trait for pioneering technologies like quantum computing and communication.

While other 2D materials are also known to emit single photons, h-BN stands out due to its numerous merits. It is considered one of the brightest single-photon sources, emitting across a broad spectral range, from infrared to ultraviolet, even at room temperature. However, there is currently a gap in comprehending the nature of defects behind single-photon emissions in h-BN, as a direct correlation between defect crystallography and emission behavior is still ambiguous in the literature¹.

We are studying samples that consist of h-BN monolayers grown on graphite (HOPG) by high-temperature molecular-beam epitaxy², using a 4 K scanning tunnelling microscope (STM) that enables directing emitted light³ into two time-resolving (30 ps) single photon detectors. Our first large-area STM-induced luminescence results confirm that these samples are optically active⁴, and high-resolution STM imaging reveals different kinds of defects on the surface. Furthermore, we aim to achieve luminescence from single defects, correlating it with their morphological and electronic properties. The combination of atomic-scale imaging, tunnelling spectroscopy and time-resolved light detection holds substantial potential for progressing the understanding of nanoscale light emission in layered materials.

References

1. S. Michaelis De Vasconcellos, D. Wigger, U. Wurstbauer, A. W. Holleitner, R. Bratschitsch, and T. Kuhn, Single-Photon Emitters in Layered Van Der Waals Materials, *Physica Status Solidi (b)* 259, 2100566 (2022).
2. T. S. Cheng, A. Summerfield, C. J. Mellor, A. Davies, A. N. Khlobystov, L. Eaves, C. T. Foxon, P. H. Beton, and S. V. Novikov, High-Temperature Molecular Beam Epitaxy of Hexagonal Boron Nitride Layers, *Journal of Vacuum Science & Technology B, Nanotechnology and Microelectronics: Materials, Processing, Measurement, and Phenomena* 36, 02D103 (2018).
3. K. Kuhnke, A. Kabakchiev, W. Stiepany, F. Zinser, R. Vogelgesang, and K. Kern, Versatile Optical Access to the Tunnel Gap in a Low-Temperature Scanning Tunneling Microscope, *Review of Scientific Instruments* 81, 113102 (2010).
4. R. J. P. Román et al., Band Gap Measurements of Monolayer H-BN and Insights into Carbon-Related Point Defects, *2D Mater.* 8, 044001 (2021).

Electronic characterization of magnetic CrCl₃ monolayers on NbSe₂

J. P. Cuperus¹, D. Vanmaekelbergh¹ and I. Swart¹

¹*Debye Institute for Nanomaterials Science, Utrecht University, Utrecht, The Netherlands*

Today's electronic devices suffer from a considerable loss of energy in the form of heat. With the ever increasing scale of information processing, it is essential to develop devices that are more efficient – either in power usage or in information processing. In both cases, new materials are required to enable this. Van der Waals heterostructures are a class of materials that are well suited to offer the required properties. By combining materials with different properties in a heterostructure with a high quality interface, a plethora of properties can be obtained. Recently, this strategy was used to obtain topological superconductivity by combining superconducting NbSe₂ with ferromagnetic CrBr₃ [1].

We have conducted STM experiments on a similar heterostructure, replacing the out-of-plane ferromagnetism of CrBr₃ by the in-plane ferromagnetism of CrCl₃. We present the MBE growth of CrCl₃ on NbSe₂ and structural insight into the formed heterostructure by STM measurements. In addition, the electronic structure is investigated using differential conductance measurements, both with and without an externally applied magnetic field. We show that the superconductivity of NbSe₂ couples to the in-plane ferromagnet, thereby introducing YSR states.

References

- [1] S. Kezilebieke, M.N. Huda, V. Vaňo, *et al. Nature* **588**, 424–428 (2020).

Tuning the planarity of a thiantherene-based molecule on Au(111)

Kwan Ho Au-Yeung¹, Suchetana Sarkar¹, Sattwick Haldar², Pranjit Das¹, Dmitry A. Ryndyk^{3,4}, Andreas Schneemann², Francesca Moresco¹

1Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany

2Inorganic Chemistry I, TU Dresden, 01069 Dresden, Germany

3Institute for Materials Science, TU Dresden, 01062 Dresden, Germany

4Theoretical Chemistry, TU Dresden, 01062 Dresden, Germany

We conducted a comprehensive study on the adsorption geometries of a non-planar aromatic thianthrene-based molecule on the Au(111) surface by scanning tunneling microscopy. The molecules demonstrate two self-assembled structures depending on the orientation of the C-S-C bonds: close-packed islands where they are non-planar and quasi one-dimensional chains where they are planar. Using the STM tip to isolate a molecule from the island, it is observed that the non-planar molecules transform into the planar configuration.

Interaction of single 4f metal-organic molecules with light

V. Rai¹, L. Gerhard¹, Q. Sun¹, C. Holzer², T. Repän³, M. Krstic², L. Yang³, A. Ebert¹, M. Wegener³, C. Rockstuhl², and W. Wulfhekel¹

¹*Institute for Quantum Materials and Technology, Karlsruhe Institute of Technology,*

²*Institute for Theoretical Solid State Physics, Karlsruhe Institute of Technology,*

³*Institute of Nanotechnology, Karlsruhe Institute of Technology.*

Single molecule light emitting sources suffer fluorescence quenching once connected to metallic leads. Therefore, to emit light, they need to be decoupled from the electrodes [1] to prevent fluorescence quenching, however, to conduct, the molecular orbitals need to hybridize with the electrodes. 4f metal-organic complexes have an immense advantage in comparison to purely organic chromophores or chromophores based on d-electrons, because the 4f states are highly localized hampering non-radiative recombination leading to long lifetimes of the excited states. 4f metal-organic complexes combine the extremely sharp emission lines including spin-orbit coupling and hyperfine coupling with functionalization by the ligands to enhance the coupling to the photon field. In future opto-electronic quantum devices the complexes can act as single photon emitters and their up- and down-conversion can be used to couple photons of different wavelength. Here, we plan to study light emitting 4f metal-organic complexes in a low temperature, ultra-high vacuum scanning tunnelling microscope (STM) [2]. While the system has been successfully used to couple out light emitted from single molecules [3] [4], it's also suitable to couple in light using the same optical path. The addition of a pulsed light source for coupling light into the STM junction will greatly extend the possibilities to study the exciton dynamics

References

- [1] J. Repp et al., Phys. Rev. Lett. 94, 026803 (2005)
- [2] K. Edelmann et al., Rev. Sci. Instrum. 89, 123107 (2018)
- [3] B. Doppagne et al. Science 361, 251-255 (2018)
- [4] V. Rai et al., Nano Lett. 10, 7600–7605 (2020)

A scanning tunnelling microscope combined with radio frequency to detect magnetic resonance at the atomic scale

Eugenio Gambari¹, Pascal David¹, Mathieu Bernard¹, François Debontridder¹, Christophe Brun¹, Tristan Cren¹, Marie Hervé¹

¹ *Institut des Nanosciences de Paris, Sorbonne Université and CNRS-UMR7588, Paris, France*

In condensed matter systems, the investigation of low energy magnetic excitations has been usually carried out using resonance methods [1]. With the development of low dimension electronics and quantum computing, one challenge in condensed matter physics is to develop resonance experiments capable of detecting low energy electronic, magnetic and vibronic excitations down to the single atom / molecule limit. Scanning tunnelling microscopy (STM) can be used to investigate these excitations of single molecules or atoms on surfaces in the sub-meV range [2]. Indeed, such excitation spectrum can be probed using conventional STM if the energy separation between the states is larger than $3.2k_B T$.

To go beyond the thermally limited energy resolution, one possibility is to combine resonance techniques and STM. In this way, the excitation spectra of single atoms and molecules can be investigated with a neV energy resolution. Accessing this region of the energy spectrum with a STM has been a long-standing problem because of its natural inability to detect high frequency signals. Only recently, some extensions of STM have been developed to detect low energy magnetic excitations down to the sub- μ eV range, allowing to perform resonance measurements such as FMR [3].

We are currently building a RF-STM setup at the Institut des NanoSciences de Paris (INSP). In this communication, I will present our current progress in the realization of this project, introducing at first the home-built experimental setup and its working principles.

References

- [1] C. P. Slichter. Principles of Magnetic Resonance (1996).
- [2] A. J. Heinrich, J. A. Gupta, C. P. Lutz, and D. M. Eigler, Science 306, 466 (2004).
- [3] M. Hervé, M. Peter, T. Balashov and W. Wulfhekel. Towards Laterally Resolved Ferromagnetic Resonance with Spin-Polarized Scanning Tunneling Microscopy. Nanomaterials, MDPI, 9 (6), pp.827 (2019).

An electrical molecular motor driven by angular momentum transfer

J. Skolaut¹, S. Marek², N. Balzer³, J. Wilhelm⁴, R. Korytár², M. Valásek³, L. Gerhard¹, M. Mayor^{3,5}, W. Wulfhekel¹ and F. Evers⁴

¹*Institute for Quantum Materials and Technology,*

Karlsruhe Institute of Technology, 76021 Karlsruhe, Germany

²*Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University, Ke Karlovu 5, 121 16, Praha 2, Czech Republic*

³*Institute of Nanotechnology,*

Karlsruhe Institute of Technology, 76021 Karlsruhe, Germany

⁴*Institute of Theoretical Physics and Regensburg Center for Ultrafast Nanoscopy, University of Regensburg, 93053 Regensburg, Germany*

⁵*Department of Chemistry,*

University of Basel, 4056 Basel, Switzerland

Inducing unidirectional motion in molecular machines inevitably requires breaking of time reversal symmetry and dissipation of energy. Observation of individual movements of single molecules in a Scanning Tunneling Microscope (STM) becomes possible when the motion of a single molecule can be broken down into a sequence of (stochastic) transitions between comparatively long-lived states. In combination, unidirectional motion is typically achieved by excitation of a molecule and a subsequent relaxation back to a rotated or translated ground state (ratchet motor). Here, we showcase a fundamentally different principle mechanism for the generation of unidirectional rotational motion of a molecule. Density functional theory (DFT) based transport calculations show that directionality can be induced by the transfer of orbital angular momentum from the tunneling current to a molecular rotator group due to chiral orbitals in the current path. In this way, the the resulting winding sense is independent of the sign of the current direction. Our molecular motors are mounted on a Au(111) surface by three anchoring groups and arranged in specific ordered structures, such that the chiral group is free to rotate while a tunneling current can be injected by the tip of the STM. Our analysis of the molecular rotational switching between three different tunneling current levels shows a statistically significant preferred direction which is indeed independent of the polarity of the applied voltage, that is the direction of the current. In full agreement with the DFT calculations, the effect is particularly strong for specific values of the applied voltage, that is for tunneling into specific orbitals.

Development of a closed-cycle dilution refrigerator scanning tunneling microscope

**Máté Stark¹, Daria Sostina², Wantong Huang¹, Paul Greule¹,
Christoph Sürgers¹, Philip Willke¹, Wolfgang Wernsdorfer¹**

¹Physikalisches Institute (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany

²Institute of Quantum Materials and Technologies (IQMT), Karlsruhe Institute of Technology, Karlsruhe, Germany

Over the past few decades, scanning probe techniques, particularly scanning tunneling microscopy (STM), have played a crucial role in advancing our understanding of atomic-scale physics. However, recent advancements in the investigation of the electronic and magnetic properties of single atoms have revealed certain experimental limitations. For example, when electron spin resonance (ESR) is combined with STM, it has been observed that the spin relaxation time (T_1) and phase coherence time (T_2) for single atoms are relatively short, primarily due to the influence of thermally excited electrons [1].

In this context, we introduce a novel approach involving the implementation of a compact closed-cycle dilution refrigerator (DR) capable of reaching milli-Kelvin temperatures. This DR is combined with a commercial STM operating under ultra-high vacuum (UHV) conditions. This combination offers several advantages, including rapid cooling, minimal vibration, and reduced routine maintenance requirements. To demonstrate the capabilities of this DR-STM system, we conducted ESR measurements on individual magnetic molecules.

References

- [1] Willke et al., Sci. Adv. 4(2), 1543 (2018)

Sample preparation in UHV for STM measurements

Caroline Hommel

Ewha Womans University, Center for Quantum Nanoscience, Seoul, South Korea

Lanthanide single molecule magnets are promising materials for the future spintronics applications, such as ultrahigh density memory devices. Using several methods, we have succeeded in synthesizing various interesting compounds, like Ln-phthalocyanine double-deckers (Ln-Pc₂; Ln = Y, Er, Tb), and endohedral fullerenes. We then deposited them on a metal substrate in ultra-high vacuum (UHV) for scanning tunneling microscopy (STM) analysis. Based on the very well-known chemical properties of the metal organic complexes, it is possible to functionalize them for various purposes. For example, Ln-Pc₂ forms ordered structures on the surface, which could be further enhanced by long chain alkane groups on the Phthalocyanine ligands. However, such large organic molecules cannot be sublimated without fragmentation. Using electro spray deposition (ESD), thin films of thermally unstable molecules can be deposited directly in UHV. Various parameters, such as pressure, time, and solvent play a role in this process. The influence of the parameters on the deposition is being investigated for different organic compounds of interest.

Reactivity of single metal atoms in different coordination environments

Longfeng Huang,¹ Nan Cao,¹ Johannes V. Barth,^{1*} Alexander Riss^{1*}

¹ Physics Department E20, Technical University of Munich, 85748 Garching, Germany

Metal centers within metalorganic networks hold promise for applications in single-atom catalysis, gas storage and sensing. Their activity is not only determined by the chemical nature of the metal, but is also significantly influenced by the bonding configuration. It is of fundamental scientific and technological relevance to understand how the reactivity of these sites can be controlled via chemical design.

In this work we present direct measurements of the interaction forces of single metal atoms^{1,2} in different metalorganic coordination environments via atomic-scale atomic force microscopy (AFM) experiments. In particular, we examine how the interaction of metal sites with different types of probe atoms (that are directly attached to the AFM tip) changes as a function of their bonding configuration. Our measurements show that while chemically rather inert CO-functionalization yields little discrimination between the metal centers in different environments, Cl-functionalization gives rise to significant differences: low-coordinated metal atoms exhibit notably stronger interactions with the probe atom.

Such atomic-scale studies provide means to characterize active sites within complex metal-organic architectures, thus aiding in a rational design strategy towards materials with tailored catalytic properties.

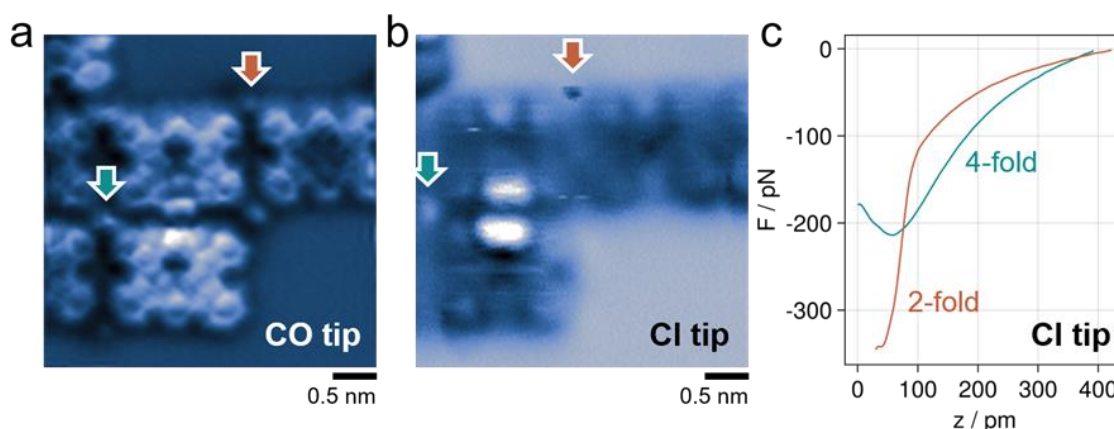


Figure 1. Interaction of Fe atoms at different sites in metalorganic networks with functionalized AFM tips. Constant height AFM images acquired with (a) CO- and (b) Cl-functionalized tips. (c) This behavior is reflected in the force-versus-distance curves: the curve taken at the twofold coordinated site (orange color) shows a substantially deeper local force minimum (i.e., stronger attraction) compared to the fourfold coordinated site (green color).

References

- [1] Huber, F. *et al. Science* **366**, 235 (2019).
- [2] Berwanger, J. *et al. Phys. Rev. Lett.* **124**, 096001 (2020).

SPIN ENGINEERING IN ARTIFICIAL ATOM-MOLECULE HYBRIDS

**Wantong Huang¹, Máté Stark¹, Paul Greule¹, Daria Sostina²,
Constantin Weeber¹, Jose Gálvez³, Christoph Sürgers¹, Christoph
Wolf³, Wolfgang Wernsdorfer^{1,2}, and Philip Willke¹**

¹*Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany*

²*Institute of Quantum Materials and Technologies (IQMT), Karlsruhe Institute of
Technology, Karlsruhe, Germany*

³*Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic
of Korea*

The combination of electron spin resonance (ESR) and scanning tunneling microscopy (STM) allows to address single spins on surfaces with unprecedented energy resolution and to control them coherently [1]. For this technique, both individual atoms as well as magnetic molecules [2,3], consisting of one central spin surrounded by ligands, are promising building blocks and offer a route for nanoscale spintronics and future quantum devices. Here, we explore the magnetic properties and spin dynamics of artificially built hybrids of atomic and molecular spins. We construct atomic dimers consisting of iron phthalocyanine (FePc) molecules and Fe atoms adsorbed on magnesium oxide by STM tip manipulation. We investigate their emergent magnetic properties by both scanning tunneling spectroscopy and ESR-STM. We believe, that such magnetic prototype structures provide a versatile platform to study complex magnetic interactions and atomic-scale spin dynamics.

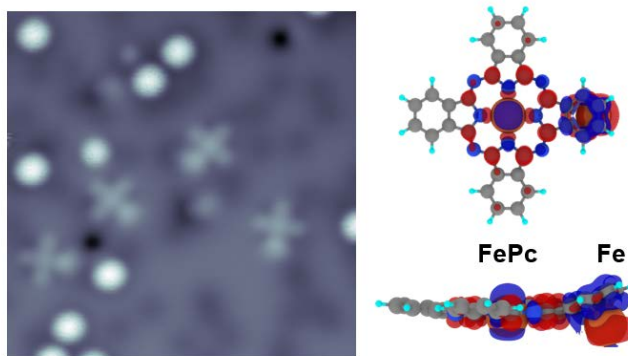


Figure 1: Left: STM topography image (9 nm x 9 nm) of four artificially built dimers of Fe and FePc using the STM tip. Right: DFT calculations of the geometry arrangement in the dimer.

References

- [1] Chen, Y., Bae, Y., A.J., et al., *Adv. Mat.*, 2107534 (2022)
- [2] Zhang, X., Wolf, et al., *Nat. Chem.*, 14(1), 59-65 (2022)
- [3] Willke, P., Bilgeri, T., et al., *ACS nano*, 15(11), 17859-17965 (2021)

Rendering a doped Mott insulator in a NbSe₂ heterostructure

**Xin Huang¹, Somesh Ganguli¹, Jiangxu Li²,
Jani Sainio¹, Jose Lado¹, Peter Liljeroth¹**

¹*Department of Applied Physics, Aalto University, Finland*

²*Department of Physics & Astronomy, The University of Tennessee, USA*

Doped Mott insulator is one of the paradigmatic models for strongly correlated systems and is a well-known platform for unconventional high-temperature superconductors like cuprates. One key question inside cuprates is how the Mott gap and emergent states around the Fermi level evolve with doping. However, the real-space study is still challenging since the cuprates contain many different elements and defects create strong chemical reconstructions in the material. Here we use heterostructure technique to render a doped Mott insulator in a transition metal dichalcogenide, via epitaxy monolayer 1T-NbSe₂ on a metallic 1H-NbSe₂ monolayer acting as a charge reservoir substrate. We use low-temperature scanning tunneling microscopy and spectroscopy to directly visualize its evolution from half-filled to the full-filled case at the atomic scale. We observe as the doping increased, this intrinsic doped Mott insulator's upper Hubbard band moves towards the Fermi level, and the lower Hubbard band shifts away from the Fermi level and gradually loses its spectral weight. The overall system has 1/3 closed to half-filled and 2/3 close to fully-filled, and hence is an over-doped scenario. Our designer artificial heterostructure provides a new matrix to tune doped Hubbard models, and a defect-free approach to explore correlated doped systems.

xin.huang@aalto.fi

A Universal Model for STM-Induced Luminescence

Song Jiang,¹ Tomáš Neuman,² Rémi Bretel,² Alex Boeglin,¹

Fabrice Scheurer,¹ Eric Le Moal,² and Guillaume Schull¹

¹ *Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France*

² *Institut des Sciences Moléculaires d'Orsay (ISMO), UMR 8214, CNRS, Université Paris-Saclay, 91405 Orsay Cedex, France*

Fluorescence of neutral and charged molecules has been reported with subnanometer resolution in scanning tunneling microscope induced-luminescence (STML) experiments over the last years.[1] Despite these significant developments, the detailed mechanisms of STML of individual molecules remained to be fully understood. In these studies, an ultrathin decoupling layer is introduced between the molecules and the supporting metal sample to suppress the substrate-induced fluorescence quenching. This strategic approach has also been employed for precise control of the charge states of single molecules,[2] which suggests that different charge states should be considered in STML process.

Here, we proposed a “universal” fluorescence model relying on successive carrier tunneling events that is based on a systematic experimental STML study of quinacridone (QA) molecules adsorbed on four-monolayer (4 ML) NaCl/Ag(111).[3] A rare property of this molecule is that neutral and positively charged fluorescence dipoles are oriented at $\approx 65^\circ$ from each other. This feature allows us to unambiguously distinguish the charged and neutral emissions solely from their distinct patterns in STML maps.

Based on a comparison between the spatial, bias voltage, and tunneling current dependences of the charged and neutral emission, and on conductance data, a many-body description of the system is set. It reveals that four different charged states of QA (QA^- , QA^0 , QA^+ , and QA^{2+}), involving three different spin multiplicities, can be populated within a single voltage sweep. This excitation mechanism is backed up by data obtained for QA deposited on NaCl/Au(111) where the ground state of the molecule is a doubly positively charged singlet state (S^{2+}). This model bears a universal character that can be applied to STML experiments dealing with charged and neutral molecules.

References

- [1] X. H. Qiu, et al., *Science* **299**, 542-546 (2003) ; Y. Zhang, et al., *Nature* **531**, 623-627 (2016) ; H. Imada et al., *Nature* **538**, 364-367 (2016) ; B. Doppagne et al., *Science* **361**, 251-255 (2018).
- [2] P. Scheurer, et al., *Phys. Rev. Lett.* **123**, 066001 (2019). K. Kaiser, et al., *Nat. Commun.* **14**, 4988 (2023).
- [3] S. Jiang, et al., *Phys. Rev. Lett.* **130**, 126202 (2023)

Creation of graphene quantum dot nanoarrays in van der Waals heterostructure

Keda Jin^{1,2,3}, Junting Zhao^{1,2,3}, Tobias Wichmann^{1,2,4}, Jose Martinez-Castro^{1,2,3}, F. Stefan Tautz^{1,2,4}, Felix Lüpke^{1,2}, Markus Ternes^{1,2,3}

1. Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany
2. Jülich Aachen Research Alliance, Fundamentals of Future Information Technology, 52425 Jülich, Germany
3. Institute for Experimental Physics II B, RWTH Aachen, 52074 Aachen, Germany
4. Institute for Experimental Physics IV A, RWTH Aachen, 52074 Aachen, Germany

jin@physik.rwth-aachen.de

Abstract

Quantum dot nanoarrays on graphene have generated interest for their ability to mimic the moiré potential observed in twistronics. While the moiré potential is sensitive to the twist angle, we present a method to periodically modulate the graphene potential by stacking graphene on 1T/2H-NbSe₂. The doping effect from the charge density wave of 1T-NbSe₂ allows for the formation of quantum dot nanoarrays independent of the periodically twist angle. Using scanning tunneling spectroscopy, we visualized the atomic-scale electronic structure of graphene/1T/2H-NbSe₂ samples with two different twist angles. These nanoarrays consistently follow the 1T-NbSe₂ charge density wave superlattice, independent of the twist angle. Furthermore, the interplay between the moiré potential and the quantum dot nanoarrays reveals novel phenomena: a larger twist showed a triangular mesh superlattice of unbounded states, while a smaller angle showed stripe patterns, breaking the C_{6v} symmetry. By varying the coupling strength between the tip and the sample, we find that the lifetime of electron in the unbound states is longer than in the bound states. Our research paves the way for the fabrication and manipulation of quantum dots nanoarrays, extending beyond graphene to other 2D van der Waals heterostructure that are not limited by twist angle.

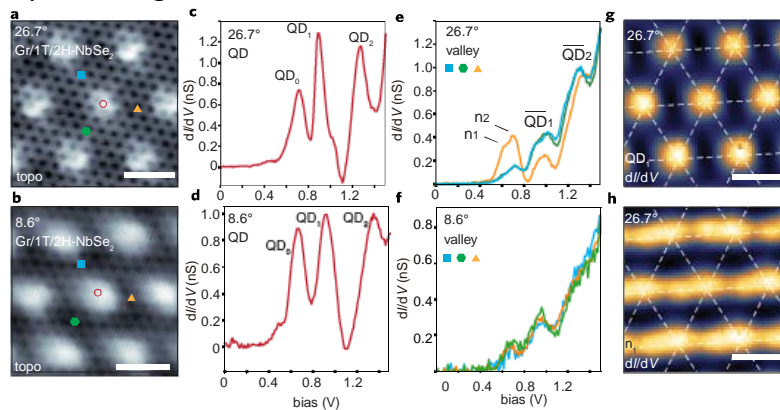


Figure 1: **a,b** topography of graphene on 1T/2H-NbSe₂ with various twist angle; **c,d** dI/dV spectra acquired on the QD; **e,f** dI/dV acquired on the valley positions as indicated in **a** and **b** respectively; **g, h** dI/dV grids of Gr/1T/2H-NbSe₂ with twist angle of 26.7° (scale bar = 1nm).

Strain-Induced Edge States on Bilayer SnTe

Liwei Jing,¹ Mohammad Amini,² Orlando J. Silveira,² Adolfo O. Fumega,² Jose L. Lado,² Peter Liljeroth,² and Shawulienu Kezilebieke^{1,3}

¹*Department of Physics, University of Jyväskylä, FI-40014 Jyväskylä, Finland*

²*Department of Applied Physics, Aalto University, 00076 Aalto, Finland*

³*Department of Chemistry and Nanoscience Center, University of Jyväskylä, FI-40014 Jyväskylä, Finland*

Topological insulators host gapless metallic states at their edges or surfaces protected by time-reversal symmetry, where the strong spin-orbit coupling induces a band inversion. So far, the concept of topology has been extended to a new phase of matter, topological crystalline insulators (TCIs), whose boundary states are protected by crystal symmetries. The new topological invariant is called the mirror Chern number rather than a topological invariant Z_2 in TIs. TCIs have been predicted and observed first in IV-VI semiconductor tin telluride^[1] (SnTe) and related alloys^[2]. However, to date, all the theoretical calculations and experiments only focus the research of TCIs on thin films or bulk materials. Here, we use molecular-beam epitaxy to grow two-dimensional islands of SnTe on superconducting niobium diselenide (NbSe₂). We then use low-temperature scanning tunnelling microscopy and spectroscopy to reveal robust edge states propagate along the sharp edges of bilayer SnTe grown on 2H-NbSe₂. The edge states shift periodically in energy across dislocation stripes, which may arise from varying strain fields due to a large mismatch between two different lattices. At low-symmetry edges, a band gap is opened. Besides, we observe the interacting edge states between adjacent islands by constant current dI/dV maps under a series of energies. This work may pave the way for interacting topological edge states under strain.

References

- [1] Junwei Liu, Timothy H. Hsieh, Peng Wei, Wenhui Duan, Jagadeesh Moodera & Liang Fu, *Nature Materials* **13**, 178-183 (2014)
- [2] Yoshinori Okada, Maksym Serbyn, Hsin Lin, et.al., *Science* **341**, 1496-1499 (2013)

Quantum Phase transitions of spin-chains on superconductors

Divya Jyoti^{1,2}, Deung-Jang, Choi^{1,2,3} and Nicolas Lorente^{1,2}

¹*Centro de Fisica de Materiales, CFM/MPC(CSIS-UPV/EHU), Paseo Manuel de Lardizabal 5, 20018, San Sebastian, Spain*

²*Donostia International Physics Center (DIPC), Paseo Manuel de Lardizabal 4, 20018, San Sebastian, Spain*

³*Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain*

Superconductivity is strongly influenced in the vicinity of a magnetic impurity or chains of magnetic atoms. Using a self-consistent approach^[1], we study the effects of a modified superconducting gap (Δ) function on the local density of states for a system of magnetic atoms placed on a 2-dimensional superconducting lattice. By solving a set of Bogoliubov-de-Gennes^[2] (BdG) equations using Nambu formalism and Green's function method, we are able to evaluate the mean-field value of the gap function. This allows us to express the effect of the magnetic impurity via a self-energy term that contains effect of scattering off the impurity via its potential part (non-magnetic) and its exchange part (magnetic). When the magnetic part is increased beyond a critical value, a quantum phase transition^[3] (QPT) marked by an abrupt drop in the value and a sign change of the superconducting gap is observed. Extension to k-space has allowed us to study an infinite chain of impurities on an infinite superconducting lattice. This led us to the investigation of Majorana bound states^[4] (MBS) which are theoretically predicted to exist in a topological regime where long atomic chains of magnetic impurities are involved. From our work, we justify that eventhough self-consistency does help us understand various phenomena, however, it shows no significant effect on the electronic density of states thereby eliminating its signature from experimental findings.

References

- [1] M. E. Flatté and J. M. Byers, Phys. Rev. Lett. 78, 3761 (1997).
- [2] Jian-Xin Zhu. Bogoliubov-de gennes method & its applications, Springer (2016).
- [3] M. I. Salkola, A. V. Balatsky, and J. R. Schrieffer, Phys. Rev. B 55, 12648 (1997).
- [4] M. H. Christensen, M. Schechter, K. Flensberg, B. M. Andersen, and J. Paaske, Phys. Rev. B 94, 144509 (2016).

Scanning Near-Field Optical Microscopy study of Metal-Insulator

Transition dynamics in VO₂/TiO₂(110) thin film

Kyungmin Kim^{1}, Linfeng Hou¹, Jun Nishida², Takashi Kumagai², Masayuki Abe¹*

¹Graduate School of Engineering Science, Osaka University

²Center for Mesoscopic Sciences, Institute for Molecular Science

*E-mail: u764704g@ecs.osaka-u.ac.jp

Vanadium dioxide (VO₂) thin films exhibit a metal-insulator transition (MIT) with a change in resistance of three orders of magnitude nearby room temperature (~340 K). The resistive transition in VO₂ is accompanied by a structural change from a low-temperature semiconducting monoclinic phase to a high-temperature metallic tetragonal rutile phase^[1]. Still, there's ongoing debate about the MIT's starting process and its progression mechanism, with in-depth research employing surface scientific methods like C-AFM^[2] and Raman spectroscopy^[3].

In this study, we grew VO₂ thin films on TiO₂(110) substrates with step and terrace structures to investigate their crystallinity and MIT dynamics in real space, by using X-Ray Diffraction (XRD) and scattering-type Scanning Near-field Optical Microscopy (s-SNOM) with Atomic Force Microscopy (AFM) simultaneously. VO₂ thin films were grown by pulsed laser deposition under the substrate temperature, the laser frequency, and the partial oxygen pressure of 723 K, 2 Hz, and 0.95 Pa, respectively. The crystallinity was characterized using X-ray diffractometer while heating and cooling the sample between 60 °C ~ 120 °C.

As a result, a noticeable shift in the VO₂(220) diffraction peak was observed (**Fig.1(a)**), which can be attributed to the structural transition from the insulator phase (at lower temperatures) to the metallic phase (at higher temperatures). Specifically, the pronounced (220) diffraction peak exhibited an angle of $2\theta = 57.56^\circ$ at 60°C, while this angle shifted to $2\theta = 57.32^\circ$ at 120°C. Regarding the AFM/s-SNOM measurements, we succeeded to visualize the dynamics of the VO₂ metal-insulator transition in real space (**Fig.1(b)**). During the presentation, we will delve into the process of MIT on the surface, by focusing on series of temperature-dependent s-SNOM images, coupled with pertinent morphological information obtained through tapping-mode AFM.

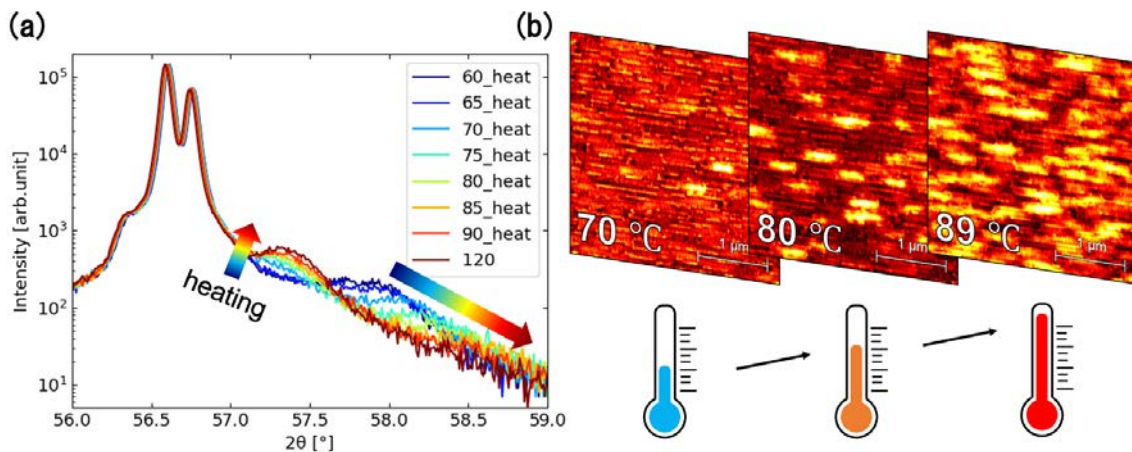


Fig 1.(a) The temperature dependence of X-Ray Diffraction spectra of VO₂ thin film grown on TiO₂(110) substrates for the heating process from 60 °C to 120 °C. **(b)** Temperature-dependent IR s-SNOM images visualizing MIT dynamics on VO₂(110).

References:

- [1] F. J. Morin, *Phys. Rev. Lett.* **3**, 34 (1959).
- [2] T. Kikuzuki et al., *Phys. Rev. B* **82**, 144113 (2010).
- [3] C. Zhang et al., *J. Mater. Sci.* **56**, 260-268 (2021).

Quantum simulator to emulate lower dimensional molecular structure

E. Sierda¹, A.M.H. Krieg¹, X. Huang¹, D. Badrtdinov¹, B. Kiraly^{1,2}, E. Knol¹, G. Groenenboom¹, M.I. Katsnelson¹, M. Rösner¹, D. Wegner¹ and A.A. Khajetoorians¹

¹ *Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands*

² *School of Physics and Astronomy, University of Nottingham, Nottingham, United Kingdom*

Designing materials with tailored physical and chemical properties requires a quantitative understanding of interacting quantum systems. In order to provide predictability, a promising route is to create bottom-up platforms, where the electronic properties of individual and interacting atoms can be emulated in a tuneable manner. Here, we present a solid state quantum simulator based solely on patterned Cs atoms on the surface of semiconducting InSb(110), a system characterized by a dilute two-dimensional electron gas decoupled from the substrate's bulk bands. We use this platform to create electron traps that emulate artificial atoms by precisely positioning Cs atoms using atom manipulation in scanning tunneling microscopy (STM). Localized states of such artificial atom are expected based on *ab initio* calculations and confirmed by probing with scanning tunnelling spectroscopy (STS). These artificial atoms serve as building blocks to realize artificial molecular structures with different orbital symmetries which are probed by spatially dependent tunnelling conductance maps. We find bonding and anti-bonding states for coupled dimers of artificial atoms and orbitals of higher symmetries (π orbitals) for a linear assembly of a few artificial atoms. Based on these artificial orbitals and various atomic patterns, we emulate the structure and orbital landscape of well known planar organic molecules, including antiaromatic molecules. In addition to this, we will show new perspectives toward simulating many-body states with this platform. Presented results validate this new quantum simulator platform, prove its high tunability. [1]

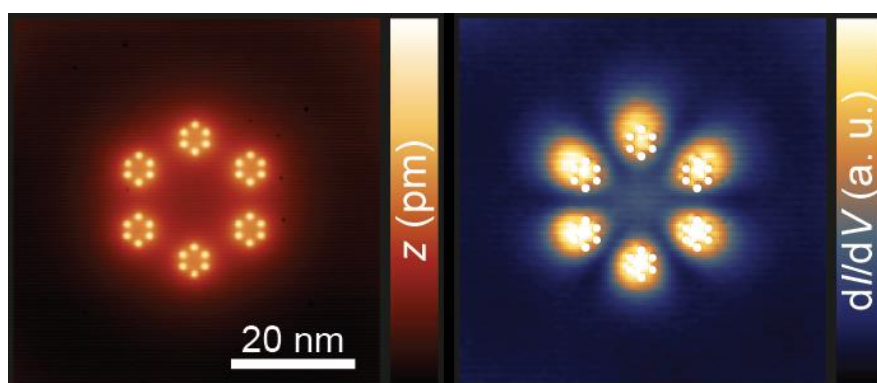


Fig. 1. STM image of artificial atoms in hexagonal arrangement resembling benzene structure and spatial distribution of one of its orbitals probed in the experiment.

[1] Sierda, E. *et al.*, *Science* 380, 1048-1052 (2023) DOI: 10.1126/science.adf2685

Measuring Yu-Shiba-Rusinov state dynamics using Stochastic Resonance Spectroscopy

Vivek Krishnakumar Rajathilakam¹, Nicolaj Betz¹, Susanne Baumann¹, Sebastian Loth¹

¹ University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

Email: vivek.krishnakumar-rajathilakam@fmq.uni-stuttgart.de

Impurities can critically influence and alter the properties of substances in many ways, giving rise to interesting new phenomena. Examples of such are dopants that introduce bound states within the bandgap in semiconductors, heavily altering their electronic properties, or magnetic impurities in metals that give rise to the Kondo effect. Similar to this, magnetic impurities coupled to a superconductor give rise to Yu-Shiba-Rusinov (YSR) states [1-3]. These are localized single-electron states in the superconducting energy gap that arise due to the interaction between the localized magnetic moments and the superconducting Cooper pairs. A Scanning Tunneling Microscope (STM) provides a great tool to engineer, manipulate and tunnel into these YSR states. However, thus far, probing the dynamics of these states has only been achieved indirectly via sequential tunneling rates [4-5]. Here, we demonstrate a more direct measurement of the YSR state dynamics using Stochastic Resonance Spectroscopy [6], which makes use of the synchronization between statistical switching and an external periodic drive. We measure the stochastic YSR state dynamics of iron (Fe) atoms coupled to a superconducting vanadium (V(001)) surface in the gigahertz range. Such direct measurement and characterization of the YSR state dynamics could open avenues for creating qubits and other quantum devices based on these states in the future.

References:

- [1] L.Yu. *Acta Phys. Sin.*, **21**(1), 75-91 (1965)
- [2] H.Shiba. *Prog. Theor. Phys.*, **40**(3), 435-451 (1968)
- [3] A. I. Rusinov. *JETP Lett.*, **9**(2), 85 (1969)
- [4] M. Ruby, et al. *Phys. Rev. Lett.* **115**, 087001 (2015)
- [5] Huang, H., et al. *Nat. Phys.* **16**, 1227-1231 (2020)
- [6] M. Hänze, et al. *Sci. Adv.* **7** eabg2616 (2021)

Huge doping and unconventional superconductivity in NbSe₂ misfit compound

Raphaël T. Leriche¹, Hugo Le Du¹, Alexandra Palacio-Morales¹, Marco Campetella¹, Cesare Tresca¹, Shunsuke Sasaki², Christophe Brun¹, François Debontridder¹, Pascal David¹, Imad Arfaoui³, Ondrej Šofranko⁴, Tomas Samuely⁴, Geoffroy Kremer⁵, Claude Monney⁵, Thomas Jaouen⁵, Laurent Cario², Matteo Calandra¹, and Tristan Cren¹

¹*Institut des Nanosciences de Paris, Paris, France*

²*Institut des Matériaux Jean Rouxel, Nantes, France*

³*Monaris, Paris, France*

⁴*Center of Low Temperature physics, Kosice, Slovakia*

⁵*Fribourg Center for Nanomaterials, Fribourg, Switzerland*

In the current quest of innovative materials which combine two-dimensionality, strong spin-orbit, valley physics, superconductivity, charge density wave, quantum-spin Hall effect, the transition metal dichalcogenides (TMD) misfit materials appear as extremely promising. They are constituted by sandwiching rocksalt layers, such as LaSe, and TMD layers such as NbSe₂. A very large combination of materials is achievable by playing on the stacking. We will show that TMD misfits are a new platform that allows achieving unprecedented high doping levels in TMD materials.

By performing ARPES, STM, quasiparticle interference measurements and first principles calculations on (LaSe)_{1.14}(NbSe₂)₂ we demonstrate a charge transfer of 0.6 electrons per Nb, almost an order of magnitude larger than what can be obtained in field effect doping [1,2,3]. Superconductivity in these compounds exhibits a huge in-plane critical field which is much higher than the paramagnetic limit [2]. We attribute this enhancement to the fact that the material behaves like a stack of weakly coupled NbSe₂ layers exhibiting a very strong Ising spin-orbit coupling. Ising superconductors are expected to host complex order parameters, such as f-wave pairing, and might even be topological materials. We will show some hint of non-conventional pairing in misfit compounds.

References

- [1] *Misfit Layer Compounds: A Platform for Heavily Doped 2D Transition Metal Dichalcogenides*, Raphaël T. Leriche et al., *Advanced Functional Materials* [31, 2007706 \(2021\)](#)
- [2] *Extreme in-plane upper critical magnetic fields of heavily doped quasi-two-dimensional transition metal dichalcogenides*, P. Samuely et al., *Phys. Rev. B* [104, 224507 \(2021\)](#)
- [3] *Periodic surface modulation of (LaSe)_{1.14}(NbSe₂)₂ observed by scanning tunneling microscopy*, Ondrej Šofranko et al., *Acta Physica Polonica A*, [137, 785 \(2020\)](#)

Signatures of an amorphous liquid of impurity states in $\text{FeTe}_{0.55}\text{Se}_{0.45}$ superconductor

**Jinwon Lee^{1,2}, Doohee Cho³, Sanghun Lee³, Koen M. Bastiaans^{1,2},
Damianos Chatzopoulos¹, Jens Paaske⁴, Brian M. Andersen⁴,
Andreas Kreisel⁴, Genda Gu⁵, and Milan P. Allan¹**

¹*Leiden Institute of Physics, Leiden University, Leiden, The Netherlands*

²*Department of Quantum Nanoscience, Technische Universiteit Delft, Delft, The Netherlands*

³*Department of Physics, Yonsei University, Seoul, South Korea*

⁴*Center for Quantum Devices, Niels Bohr Institute, University of Copenhagen, Copenhagen, Denmark*

⁵*Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY, USA*

An impurity in superconductors can break the Cooper pairs into quasiparticles, having quasiparticle states within the superconducting gap [1-3]. The in-gap states, called Yu-Shiba-Rusinov (YSR) states, are of importance because their characteristics manifest the superconducting ground state and because they can have non-trivial topological properties, especially when the YSR states have a substantial spatial extent, resulting in significant overlap between them [4]. In this work, we discover signatures of an amorphous liquid phase of impurity states in the iron-based superconductor $\text{FeTe}_{0.55}\text{Se}_{0.45}$ using scanning tunneling microscopy. By observing their spatial dispersions and junction-resistance dependency, we identify the peaks in the density of states beyond the superconducting coherence peaks as impurity states instead of the superconducting coherence peaks from other bands with larger gaps. Combining our simulations with a large density of these impurity states observed in our experiments, we propose the present findings indicate an amorphous impurity-state liquid, where the impurity states lack translational symmetry but exhibit hybridization between them. This implies that the properties of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ might be different from the conventional Fermi liquid based on lattice models.

References

- [1] L. Yu, *Acta Phys. Sin.* **21**, 75-91 (1965)
- [2] H. Shiba, *Prog. Theor. Phys.* **40**, 435-451 (1968)
- [3] A. I. Rusinov, *Zh. Eksp. Teor. Fiz.* **56**, 2047-2056 (1969)
- [4] S. Nadj-Perge *et al.*, *Science* **346**, 602 (2014)

Ultrafast measurements of Charge Density Wave dynamics with a THz light-coupled STM

Mohamad Abdo^{1,2,3}, Steffen Rolf-Pissarczyk^{2,3}, Kurt Lichtenberg¹, Shaoxiang Sheng¹, Jacob A. J. Burgess^{2,4}, Luigi Malavolti^{1,2,3} and Sebastian Loth^{1,2,3}

¹*University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany.*

²*Max Planck Institute for Solid State Research, Stuttgart, Germany.*

³*Max Planck Institute for Structure and Dynamics of Matter, Hamburg, Germany.*

⁴*Department of Physics and Astronomy, University of Manitoba, Winnipeg, Canada.*

Charge-density waves (CDWs) feature collective excitations that can be observed as an oscillatory response of the material's electron system to a fast optical stimulus [1]. While on a local scale, scanning tunneling microscopy (STM) measurements reveal highly localized interactions with atomic defects [2]. In our experiment, we combine optical THz excitation spectroscopy with localized electronic probing of an STM to enable fast excitation of the CDW locally and to probe the dynamics of CDWs at the atomic scale. We report the observation of dynamics with frequencies in the range of 0.15-0.8 THz in the 2D layered material 2H-NbSe₂, excited by the tip-enhanced electric field of THz pulses. The observed dynamics are highly heterogeneous with spatial variations down to one unit cell of the CDW. Ginzburg Landau's Theory enables us to empirically model these charge density waves and their dynamics [3,4]. Specifically, we developed a field theoretical Ginzburg Landau model for CDW dynamics, with which we can attribute the nature of the observed dynamics to overlapping phase wave packets, resulting from the interaction of the electric field-driven CDW with impurity-induced pinning sites. This observation of phase dynamics of charge density waves is unprecedented and opens the door to studying collective charge-order dynamics at the intrinsic length scale of the underlying interactions.

References

- [1] M.-A. Méasson et. al, PRB **89**, 060503(R) (2014)
- [2] C. J. Arguello et. al, PRB **89**, 235115 (2014)
- [3] W. L. McMillan, PRB **12**, 1187-1196 (4) (1975)
- [4] G. Grüner, Density Waves in Solids. Perseus Publishing - Cambridge, Massachusetts, (2000)

Distance-Dependence of Orbital Density Mapping Using a CO-Functionalized STM Tip

Leonard-Alexander Lieske¹, Fabian Paschke¹, Florian Albrecht¹ and Leo Gross¹

¹ IBM Research Europe - Zurich, Rüschlikon, Switzerland

Functionalized tips are frequently used in high-resolution AFM, particularly CO terminated tips, to facilitate atomic resolution. [1] CO functionalized tips can also enhance contrast in STM measurements of ionic resonances, mapping orbital densities. [2,3] For CO tips both *s*- and *p*-wave character of the tip contribute to the contrast. [3,4,5] Here, we study the contributions of *s*- and *p*-wave character of CO tips as a function of tip-sample distance, bias voltage and tunneling current.

The appearance of frontier molecular orbital densities of pentacene on a bilayer of NaCl grown on Cu(111) was investigated using a CO-functionalized STM tip and as a function of the tip-sample distance. Constant-height STM imaging reveals a clear transition from *p*- to *s*-wave dominated tunnelling upon tip retraction over a range of 4 Å and thus four orders of magnitude of tunnelling current. The contrast is explained with the symmetry-dependent decay lengths of the CO tip-states. Calculations using Bardeen's approach inherently reproduce the distance-dependence using a fixed ratio of *s*- to *p*-wave tip states of 60:40.

References

- [1] L. Gross, F. Mohn, N. Moll, P. Liljeroth and G. Meyer, *Science*, 325(5944), 1110-1114 (2009).
- [2] J. Repp, G. Meyer, S. M. Stojković, A. Gourdon and C. Joachim, *Physical Review Letters*, 94(2) 026803 (2005).
- [3] L. Gross, N. Moll, F. Mohn, A. Curioni, G. Meyer, F. Hanke and M. Persson, *Physical Review Letters*, 107(8), 086101 (2011).
- [4] N. Pavliček, I. Swart, J. Niedenfür, G. Meyer and J. Repp, *Phys. Rev. Lett.* 110, 136101 (2013).
- [5] A. Gustafsson and M. Paulsson, *Phys. Rev. B*, 93, 115434 (2016).

Towards topological edge states with artificial electronic lattices: Cs on InAs(111)

R.A.M. Ligthart¹ and I.Swart¹

¹ *Condensed Matter and Interfaces, Debye Institute for Nanomaterials science, Utrecht, The Netherlands*

Artificial electronic lattices are a promising tool to study topology on an atomic scale. The Scanning Tunnelling Microscope (STM) allows to build the topological lattices by manipulating single atoms with nanoscale precision and to probe their electronic properties. Here, the semiconductor InAs(111)A surface with adsorbed Cs atoms is studied. Vertical and lateral manipulation allow to place multiple Cs-adatoms in vicinity of each other. Lines of positively charged Cs atoms create a potential well that confines the surface state electrons of InAs due to local band bending. The confined state acts as an artificial atom and can be used to construct dimers and other structures. [1] Artificial atoms can therefore be used to study the topological properties of the model systems such as the SSH [2] and trimer chain.

References

- [1] E. Sierda, et al. *Science* **380**, 1048-1052 (2023)
- [2] Van Dong Pham, et al. *Phys. Rev. B*, **105**, 125418 (2022)

Aging behavior in a self-induced spin glass

Lorena Niggli¹, Julian H. Strik¹, Zhengyuan Liu¹, Mikhail I. Katsnelson¹, Daniel Wegner¹, and Alexander A. Khajetoorians¹

¹*Institute of Molecules and Materials (IMM), Radboud University, Nijmegen, The Netherlands*

Elemental neodymium has been shown to be a so-called self-induced spin glass, which, unlike conventional spin glasses, shows glassy behavior solely from frustrated magnetic interactions and in the absence of disorder [1]. Additionally, in contrast to most conventional spin glasses, neodymium has a disorder to order transition. As the temperature increases it goes from the self-induced spin glass phase to a multi-Q long range ordered phase resulting from competing long-range interactions on different length scales [2]. This disorder to order transition might have profound effects on the metastable states and aging dynamics in the low temperature phase. In this study, we use spin-polarized scanning tunneling microscopy combined with varying external magnetic fields to investigate the aging dynamics in the spin glass phase, and quantify this in terms of the various local spin-spin correlations (Q-states) observed. These observations may have interesting implications on the aging behavior in self-induced spin glasses, which might differ from conventional spin glasses.

References

- [1] U. Kamber et al., *Science* **368**, 966 (2020).
- [2] B. Verlhac et al., *Nat. Phys.* **18**, 905 (2022).

Development of an Ultra High Vacuum and Low Temperature Scanning NV Magnetometer

S. Maity¹, D. Pinto¹, R. J. Peña Román¹, K. Kern^{1,2} and A. Singha¹

¹*Max Planck Institute for Solid State Research, Stuttgart, Germany*

²*Institut de Physique, École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland*

Nitrogen vacancy (NV) center is a versatile magnetic sensor to probe sensitive spin phenomena underlying magnetic structures [1, 2]. The nanoscale spatial resolution and ambient condition measurement capabilities of NV sensors have enabled us to perform magnetic imaging through scanning probe microscopy (SPM) across a wide range of temperature and pressure, making NV-SPM a very lucrative alternative to Magnetic Force Microscopy (MFM) and other magnetic imaging techniques [1]. Here I will be discussing the development of a scanning probe magnetometer which is capable of imaging magnetic nanostructures with a high spatial resolution. In this setup, NV centers are integrated within diamond tips for performing Atomic Force Microscopy (AFM) measurements. This NV-SPM is operable in ambient and also under ultra-high vacuum (UHV) and low temperature (LT) (10^{-10} mbar and 4 K) conditions, enabled with an external vector magnetic field (1 T in z and 250 mT in both x and y direction). We have used NV tips with a novel tip holder equipped with an AFM amplifier and microwave excitations on the tip (not on the sample), allowing us to have a magnetic image of any region of a sample without restriction. Optically Detected Magnetic Resonance (ODMR) using Zeeman splitting can locally quantify the stray magnetic field from a sample. Additionally, the integrated facilities involving UHV and low temperature capabilities will allow us to investigate the stability of the NV probes and the effects of surface modifications at UHV condition, in a highly controlled manner for the first time.

References

- [1] Rondin et al, Rep. Prog. Phys. **77**, 056503 (2014)
- [2] C. L. Degen, Reviews of Modern Physics **89**, 035002(2017)

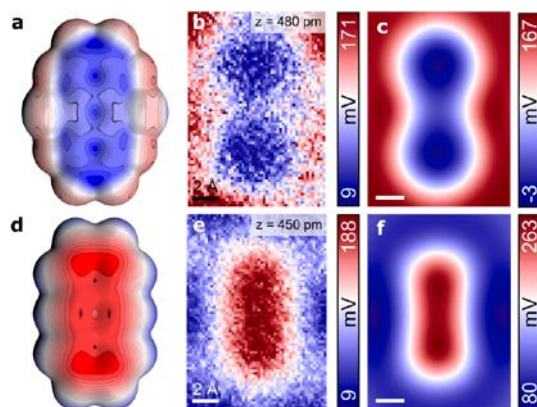
Visualization of π -hole in molecules by means of Kelvin probe force microscopy

Benjamin Mallada, Martin Ondráček, Maximilian Lamanec, Aurelio Gallardo, Alejandro Jiménez-Martín, Bruno de la Torre, Pavel Hobza, Pavel Jelínek

*Regional Centre of Advanced Technologies and Materials,
Czech Advanced Technology and Research Institute (CATRIN), Palacký University
Olomouc, 78371 Olomouc, Czech Republic.*

mallada@fzu.cz

Submolecular charge distribution significantly affects the physical-chemical properties of molecules and their mutual interaction. One example is the presence of a π -electron-deficient cavity in halogen-substituted polyaromatic hydrocarbon compounds, the so-called π -holes, the existence of which was predicted theoretically [1], but the direct experimental observation is still missing. Here we present the resolution of the π -hole on a single molecule using the Kelvin probe force microscopy [2], which supports the theoretical prediction of its existence. In addition, experimental measurements supported by theoretical calculations show the importance of π -holes in the process of adsorption of molecules on solid-state surfaces. This study expands our understanding of the π -hole systems and, at the same time, opens up possibilities for studying the influence of submolecular charge distribution on the chemical properties of molecules and their mutual interaction.



References

- [1] Politzer, P. & Murray, J. S. Electrostatics and Polarization in σ and π - Hole Noncovalent Interactions: An Overview. *ChemPhysChem* vol. **21** 579–588 (2020).
- [2] Mallada, B., Ondráček, M., Lamanec, M. et al. Visualization of π -hole in molecules by means of Kelvin probe force microscopy. *Nat Commun* **14**, 4954 (2023)

Probing the coherent amplitude mode in 1T-TaS₂ with THz scanning tunneling microscopy

L.E. Parra López¹, V. Sleziona¹, A. Vaitis¹, F. Schulz², M. Wolf¹ and M. Müller¹

¹*Fritz Haber Institute of the Max Planck Society, Berlin, Germany.*

²*CIC nanoGUNE, Donostia-San Sebastian, Spain.*

E-mail: lopez@fhi-berlin.mpg.de

Real-time spectroscopy of coherent phonons (CP) is a valuable tool in ultrafast condensed matter physics to investigate the interplay between electronic and lattice degrees of freedom in solid crystals. In strongly correlated materials, this interplay can lead to emergent quantum phenomena such as unconventional superconductivity, insulator-to-metal transitions, or the formation of charge density waves (CDWs). One of the most intriguing and widely studied quantum materials is 1T-TaS₂. At low temperatures, it exhibits a commensurate CDW phase leading to the opening of a large Mott gap [1,2]. Furthermore, at the nanoscale, inhomogeneities such as defects, impurities and nanometer-sized domains of different phases can coexist, leading to new macroscopic states. One way to investigate these states is through CP spectroscopy. It is known that the amplitude mode (AM) phonon, an ubiquitous mode to 2D materials, modulates both the Mott gap and the entire band structure of TaS₂ [3] making it an ideal probe for electron-lattice interaction. However, the spatial averaging nature of the conventional time-domain approaches is inadequate to investigate nanoscale inhomogeneities. In consequence, an experimental approach combining angstrom spatial and femtosecond temporal resolution is highly desirable.

In this work, we demonstrate that THz-induced scanning tunneling microscopy (THz-STM) can probe the coherent AM in 1T-TaS₂. To this end, we couple broadband single-cycle THz pulses to an STM junction to drive ultrafast tunneling currents, and use them to probe the ultrafast time evolution of 1T-TaS₂ in the Commensurate CDW phase after ultrafast photoexcitation with near-infrared laser pulses. We observe a 3% periodic modulation of the rectified tunneling current at a frequency of 2.5 THz, which is in good agreement with the reported value for the AM in 1T-TaS₂. Our results are an important step towards the application of THz-STM to study the nonequilibrium dynamics of quantum materials at the atomic scale.

References

- [1] B. Sipoš *et al.*, *Nature Materials* **7**, 960-965 (2008)
- [2] C. Butler *et al.*, *Nature Communications* **11**, 2477 (2020)
- [3] J. Maklar *et al.*, *arXiv:2206.03788* (2022)
- [4] S. Hellmann *et al.*, *Nature Communications* **3**, 1069 (2012)

Fusing Open-Shell Molecules on NaCl by Atom Manipulation

**F. Paschke¹, M. Vilas-Varela², S. Fernández Castro², F. Albrecht¹,
D. Peña², and L. Gross¹**

¹*IBM Research Europe, Saeumerstrasse 4, 8803 Rueschlikon, Switzerland*

²*CiQUS and Departamento de Química Orgánica, Universidade de Santiago de Compostela, 15782 Santiago de Compostela, Spain*

1D spin chains or 2D spin networks are particularly interesting not only for future spintronic applications but also to explore fundamental, albeit complex physical phenomena like many-body correlations or quantum phase transitions [1]. The generation and investigation of tailored chains and networks of various size and different subunits is thus highly desirable. This has become possible with the advent of on-surface synthesis (OSS) methods in combination with high-resolution scanning probe techniques, which allows the atomically precise synthesis and characterization of hitherto elusive molecular nanostructures [2]. However, most OSS methods rely on the catalytic activity of a metal surface, which in turn can affect essential properties of the synthesized molecule due to significant molecule-substrate interaction.

Here we use the large range of manipulation abilities provided by the tip of a scanning tunneling microscope (STM) to construct artificial molecular spin chains on an insulating layer of NaCl - molecule by molecule [3]. Brominated, nonbenzenoid truxene hydrocarbons have been chosen to serve as functional building blocks, each hosting a $S=3/2$ ground state in its pristine form [4]. The bottom-up synthesis route of extended chains comprises dehalogenation, lateral manipulation, intermolecular bond formation and subsequent dehydrogenation. Precursor molecules, reaction intermediates and resulting spin chain nanostructures are identified by means of high-resolution atomic force microscopy (AFM). Orbital density mapping of the frontier molecular states serve as starting point to determine the spin ground state by comparison to first-principle calculations.

References

- [1] D.-J. Choi et al., *Rev. Mod. Phys.* **91**, 041001 (2019).
- [2] S. Clair et al., *Chem. Rev.* **119**, 4717 (2019).
- [3] Q. Zhong et al., *Nat. Chem.* **13**, 1133 (2021).
- [4] S. Mishra et al., *ACS Nano* **16**, 3264 (2022).

Investigating Spin Noise and Stray Magnetic Field in Synthetic Antiferromagnetic Structures using Nitrogen-Vacancy Scanning Probe Microscopy

**R. J. Peña Román¹, D. Pinto^{1,2}, S. Maity¹, F. Samad^{3,4}, O. Hellwig,^{3,4}
K. Kern,^{1,2} and A. Singha¹**

¹*Max Planck Institute for Solid State Research, Stuttgart, Germany*

²*Institut de Physique, Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland*

³*Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden, Dresden, Germany*

⁴*Institut für Physik, Technische Universität Chemnitz, Chemnitz, Germany*

Syntactic antiferromagnetic (AF) structures are interesting systems that provide insights into the physics of various magnetic structures and surfaces. In addition to their potential technological applications, these are particularly attractive for magnetic storage devices and spintronic [1,2]. The study of AF films differs from ferromagnetic (FM) ones since they only have an atomic-scale net magnetic moment. Consequently, studying such a system requires techniques like X-ray photoemission electron microscopy and spin-polarized scanning tunneling microscopy, which offer high spatial resolution and magnetic sensitivity [3]. However, these methods typically require complex synchrotron facilities, low temperatures, and conductive samples.

In nitrogen-vacancy scanning probe microscopy (NV-SPM), we combine the spatial resolution of atomic force microscopy with the exceptional magnetic sensitivity of a single NV defect in diamond as a sensor. This technique, being independent of any external perturbation (such as current, magnetic field, etc.), is the least-invasive scanning probe approach. It also allows quantitative measurements of the stray magnetic fields generated by the sample. We utilize a home-built NV-SPM setup for investigating the FM domain walls and AF domains in AF-coupled multilayer thin film samples with perpendicular magnetic anisotropy. By performing measurements with two different orientations of the NV-probe, we reveal distinct fingerprints emerging from spin noise and constant stray magnetic fields from the sample. This demonstrates that NV-SPM opens up novel opportunities for understanding magnetic thin films quantitatively and non-invasively, along with exceptionally high magnetic sensitivity at the nanoscale.

References

- [1] R. A. Duine *et al.* Nature Physics, **14**, 217 (2018)
- [2] O. Hellwig *et al.* Nature Materials, **2**, 112 (2003)
- [3] S. Wook Cheong *et al.* npj Quantum Materials, **5**, 3 (2020)

Modulation of the tunneling barrier in an Anderson impurity Hamiltonian to study ESR-STM

J. Reina-Galvez^{1,2}, Yu Jisoo^{1,3}, N. Lorente⁴ and C. Wolf^{1,2}

¹*Center for Quantum Nanoscience, Institute for Basic Science, Seoul, South Korea*

²*Ewha Womans University, Seoul, South Korea*

³*Department of Physics, Ewha Womans University, Seoul, South Korea*

⁴*Centro de Fisica de Materiales, San Sebastian, Spain*

The demonstration of reproducible single-atom and single-molecule electron spin resonance (ESR) in a scanning tunneling microscope (STM) has opened new opportunities for surface science analysis at the atomic scale. These systems can be engineered to have tailored spin states, manipulated to form structures with specific interactions, and, most recently, were utilized to implement basic quantum logic operations, such as a control-NOT gate, in a STM. [1]

In this work, we aim to achieve a reliable model for Ti on a silver substrate using 2 monolayers of magnesium-oxide as decoupling layer– the benchmark system in ESR-STM. We will reproduce features of the experiments, starting from density functional theory and our own theoretical framework. Our method combines the formalism of open quantum systems with the non-equilibrium Green's Function technique.[2] The foundation of our model is a modified Anderson impurity Hamiltonian, where a quantum impurity is coupled via hopping terms to a polarized STM tip and an isolated surface, both treated as fermionic baths. The modified Anderson impurity model encompasses localized electronic states, a Coulomb repulsion term, and a Zeeman term. The experimental application of a time-dependent AC voltage introduces a modulation of the tunneling barrier, thereby influencing the hopping terms and enabling the control and manipulation of the quantum impurity, leading to resonances in the system.

We will demonstrate how driving the system on resonance following our approach alters the populations and the calculated physical current through the many-body impurity matches some of the main experimental results.

References

- [1] Yu Wang et al., Arxiv (2022)
- [2] J. Reina-Galvez et al. PRB (2021,2023)

Atomically thin transition metal halide NiBr₂ grown on Au(111)

D. Rothhardt¹, Z. Ahmed^{2, 3}, H. Liu^{2, 3}, R. Hoffmann-Vogel¹,
H. J. Hug^{2,3}, and A. Kimouche¹

¹ Institute of Physics and Astronomy, University of Potsdam, 14476 Potsdam-Golm, Germany

² Swiss Federal Laboratories for Materials Science and Technology, CH-8600 Dübendorf, Switzerland

³ Department of Physics, University of Basel, CH-4056 Basel, Switzerland

In recent years, two-dimensional (2D) van der Waals materials have emerged as new platform for studying low-dimensional quantum phenomena. This stems mainly from confined electrons and the great in play stability provided by strong covalent bonds and relatively weak van der Waals interlayer interaction. Due to their limited symmetry, they display electrical, optical, and magnetic properties that are different compared to their 3D counterparts. As novel 2D materials, transition metal dihalides (TMHs) emerge as promising platform for stable ferro- or antiferromagnetic ordering in two dimensions [1]. Up to now only few publications show the growth of monolayer TMH [2-4] however, their local magnetic visualization is still lacking. In addition, epitaxial single layer TMHs requires an accurate control of the growth parameters and an improved understanding of the interactions between TMHs and the surface. In this work, we employ frequency-modulated scanning force microscopy in the non-contact mode, combined with Kelvin probe microscopy and scanning tunnelling microscopy, to examine various phases and layer thicknesses of NiBr₂ on Au(111) revealing a complicated growth mode in the low coverage regime.

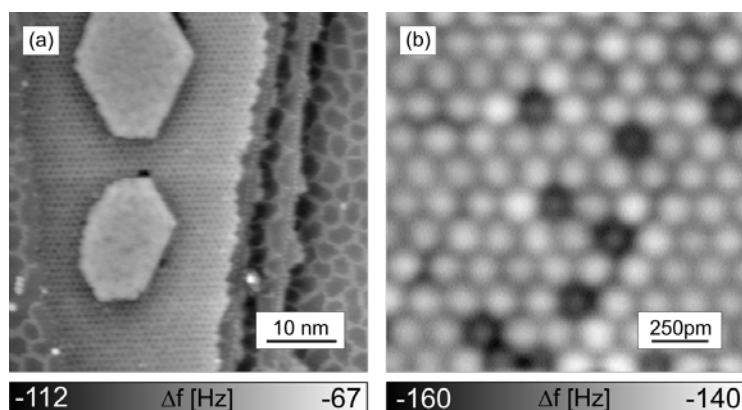


Figure 1 First flexural mode resonance frequency shift data recorded at constant tunnelling current (a) Overview ($I_t = 10$ pA, $V_{\text{bias}} = 1000$ mV) and (b) atomic resolution of the NiBr₂ ($I_t = 50$ pA, $V_{\text{bias}} = 20$ mV).

References

- [1] V.V. Kulish, *et al.*, J. Matter Chem C, **5**, 8734 (2017).
- [2] D. Bikaljević, *et al.*, ACS Nano, **15**, 14985 (2021).
- [3] H. Liu, *et al.*, ACS nano, **14**, 10544 (2020).
- [4] S.Terakawa, *et al.*, J. Phys. Chem. C, **30**, 14898 (2023).

Controlling hybridization in Yu-Shiba-Rusinov dimers through a charge-density wave

L. M. Rütten¹, E. Liebhaber¹, K. Rossnagel^{2,3}, and K. J. Franke¹

¹*Fachbereich Physik, Freie Universität Berlin, Berlin, Germany*

²*Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Kiel, Germany*

³*Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany*

Unpaired adatom spins on superconductors interact with the Cooper pairs of the substrate and induce so-called Yu-Shiba-Rusinov (YSR) states inside the superconducting gap [1-3]. These states can be probed by scanning tunneling spectroscopy at the single atom scale. In layered van der Waals materials, the YSR wave functions can extend over several nanometers [4]. Due to this large spatial extent the YSR wave functions originating from two impurities can overlap and therefore hybridize [5]. Such a hybridization can be observed as a splitting of YSR resonances in tunneling spectra. Additionally, the YSR states of magnetic adatoms can be influenced not only by the adsorption site on the crystal, but also by the presence of a charge-density wave (CDW). Fe atoms on *2H-NbSe₂* have been shown to exhibit long-range YSR wave functions, where the energies, at which the YSR resonances are observed, depend strongly on the atom's position with respect to the CDW [6].

Here, we use the tip of a scanning tunneling microscope to arrange Fe dimers on *2H-NbSe₂*. We observe, that the incommensurate CDW, that coexists with superconductivity at low temperatures in *2H-NbSe₂*, can be influenced by the positions of Fe atoms on the surface. Moving additional atoms at several nanometers distance from the dimers, we change the arrangement of the CDW with respect to the crystal lattice around our dimers. By influencing the CDW, we can make both dimer atoms reside in equivalent or non-equivalent sites with respect to the CDW. We only observe hybridized YSR states if both dimer atoms sit in equivalent CDW sites (*i.e.* exhibit YSR states at equivalent energies). Therefore, we can controllably switch YSR hybridization in our dimers on and off without directly addressing the dimer atoms.

References

- [1] L. Yu, *Acta Phys. Sin.* **21.1**, 75-91 (1965)
- [2] H. Shiba, *Prog. Theor. Phys.* **40.3**, 435-451 (1968)
- [3] A. Rusinov, *JETP Lett. (USSR)(Engl. Transl.)* **9**, 85 (1969)
- [4] G. Ménard, *et al.*, *Nature Physics* **11.12**, 1013-1016 (2015)
- [5] S. Kezilebieke, *et al.*, *Nano Letters* **18.4**, 2311-2315 (2018)
- [6] E. Liebhaber, *et al.*, *Nano Letters* **20.1**, 339-344 (2020)

Tip induced ring growth of Pb-islands on Si(111)-(7x7)

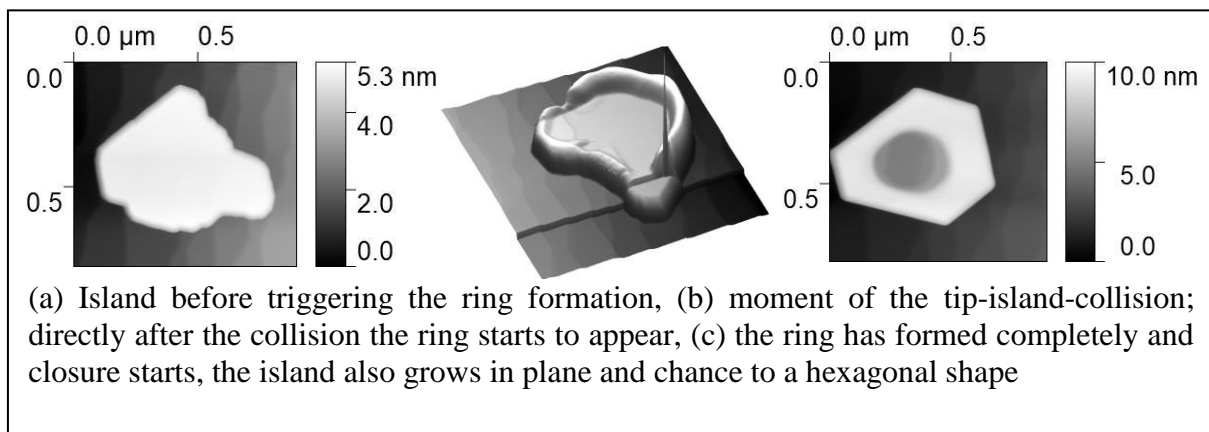
P. P. Schmidt¹, F. Hartmann¹, L. Faber¹ and R. Hoffmann-Vogel¹

¹*Institute of Physics and Astronomy, University of Potsdam, Potsdam, Germany*

Researchers have often studied the growth of Pb islands as a model system to explore the quantum size effect (QSE) [1]. Due to the QSE this system shows bilayer stability and a height growth by bilayers. The height increase occurs through ring growth [2], with rings forming at the edges of the islands and then closing inwards. In our cantilever scanning force microscopy measurements at different temperatures between 120K and 300K after evaporating Pb under ultra-high vacuum-condition (base pressure $\sim 10^{-10}$ mbar) on a cooled Si-substrate, we did not observe spontaneous ring growth. To study ring growth, we caused a collision between the scanning force microscope tip and the island. The local work function difference is determined by Kelvin probe force microscopy. With our methodology we investigate diffusion due to locally induced non-equilibrium. We show that the time scale for the formation of the ring and the closure is clearly different. In addition, the growth of the ring on the island is significantly slower compared to that in the wetting layer in laser induced thermal desorption (LITD) experiments [3]. Furthermore, data has been collected on how the centers of mass of the islands move in the xy-plane as a function of time. This data suggest a directional correlated growth.

References

- [1] T. Späth et al, *Nanoscale* **9**, 7868-7874 (2019)
- [2] M. Li et al, *Physical Review B* **79**, 113404 (2009)
- [3] L. Huang et al. *Physical Review Letters* **108**, 026101 (2012)



Atomically resolved luminescence in a Terahertz Scanning Tunneling Microscope

Johannes Schust¹, Kurt Lichtenberg¹, Felix Huber¹, Susanne Baumann¹ and Sebastian Loth¹

¹*Institute for Functional Matter and Quantum Technologies, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany*

The interplay between light and matter has a key role in solid-state research, and it constitutes a fundamental element of numerous contemporary technologies. Therefore, exploring the photonic properties of novel quantum systems is of high significance, while pushing the frontiers of both temporal and spatial resolution remains a persistent challenge. Here, we show the implementation of electroluminescence sensing, i.e. the capability to couple light directly out of the junction, into an ultra-fast Terahertz Scanning Tunneling Microscope (STM). Our exceptionally designed vacuum chamber features a thin slot that hosts the whole measuring unit, including a flow-cryostat, while quartz windows on both sides give optical access and allow for the strait-forward adjustment of all further components at ambient conditions, precluding any complicated in-vacuum optical adjustments. Multi-path optical access is easily implemented and modified to the experimental needs in that way. In the future, the concept of this setup gives the opportunity to access the junction using tunable laser radiation, that, funneled on the surface by the STM tip, will allow us to face photoluminescence measurements, too. With our unique setup, we will harness the remarkable time resolution obtained by single Terahertz voltage pulses for time-resolved electroluminescence experiments. In particular, we will unravel field-induced electroluminescence with femtosecond time resolution, while addressing samples with the atomic resolution provided by scanning probe microscopy. This allows us to access the ultra-fast dynamics of optical properties for both molecules and organic substrates.

Probing the magnetism of Tb atoms on MgO

J. Schwenk¹, S. Shan¹, C. Soulard¹, J. Dolezal¹, F. Patthey¹
and H. Brune¹

¹ *Laboratory of Nanostructures at Surfaces (LNS), Institute of Physics (IPHYS),
EPFL, Lausanne, Switzerland*

Single Tb atoms adsorbed on an MgO surface at low temperatures exhibit a stable magnetic moment. We use spin polarized STM to study the stability and magnetic properties of the species adsorbed to top oxygen and oxygen bridge sites via two-state switching time traces, STS and stochastic resonance measurements [1]. The experiments are conducted in our home built STM operating at a base temperature of 400 mK and in a vector magnetic field. A bias modulation in the STM tunnel junction required for ESR and stochastic resonance measurements is possible in a wide bandwidth from DC to 40 GHz due to a direct RF wiring of the STM tip.

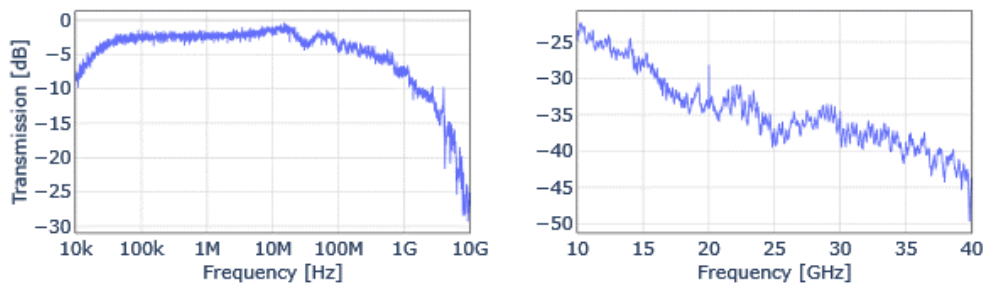


Fig. 1: Transfer function of our ESR-STM measured from 10 kHz up to 40 GHz, including the loss of 6 m RF cable in air

References

- [1] M. Hänze et al., *Sci. Adv.* **7**: eabg2616 (2021)

Excited-state spectroscopy of individual molecules with atomic force microscopy

L. Sellies¹, J. Eckrich¹ and J. Repp¹

¹*Universität Regensburg, Regensburg, Germany*

The combination of scanning probe microscopy with an increasing number of spectroscopic techniques provides unprecedented microscopic understanding. In particular, the integration of optical spectroscopy into scanning tunneling microscopy (STM) such as Raman and luminescence has recently provided breathtaking atomic-scale insight into light-matter interactions. Despite its very direct access to well-defined single-molecule model systems, the unambiguous assignment of observations to specific quantum transitions is not always straightforward [1,2].

Here, we propose a novel type of single-molecule spectroscopy that enables access to many quantum transitions of different types, including radiative, non-radiative and charge-related transitions. It builds on the recently developed technique to measure the tunneling current of single electrons between a conductive tip and a single molecule on an insulating surface [3,4]. The molecules of interest are deposited on thick insulating films, preventing tunneling of charges to and from the underlying support. By controlled charge exchange between the conductive tip of an atomic force microscope, we bring a molecule into different electronic configurations [4]. The detection proceeds via the force acting on the tip. Thereby, quantum transitions can be mapped out in energy, enabling access to the energies of the lower-lying electronic states of individual molecules, including out-of-equilibrium states. We demonstrate our technique by applying it on two different molecules.

References

- [1] K. Kimura et al., *Nature* **570**, 210-213 (2019)
- [2] J. Dolezal et al., *ACS Nano* **16**, 1082–1088 (2022)
- [3] L. L. Patera et al., *Nature* **566**, 245-248 (2019)
- [4] J. Peng et al., *Science* **373**, 452-456 (2021)
- [5] S. Fatayer et al., *Physical Review Letters* **126**, 176801 (2021)

Defect identification of Nitrogen-doped graphene on Pt (111) using AFM/STM

Hyunmin Kang,^{1,†} Jeong Ah Seo,^{2,3,†} Taegeun Yoon,^{1,†} Yongtae Cha,⁴
Jiwon Park,⁵ Bong Gyu Shin,¹ Andreas J. Heinrich,^{2, 3*} Hyo Won
Kim,^{6*} Jungseok Chae,^{2, 7*} and Young Jae Song^{1, 8, 9*}

¹ SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon 16419, Republic of Korea

² Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul 03760, Republic of Korea

³ Department of Physics, Ewha Womans University, Seoul 03760, Republic of South Korea

⁴ Department of Chemical Engineering, Sungkyunkwan University, Suwon 16419, Republic of Korea

⁵ Department of Chemistry, Sungkyunkwan University, Suwon 16419, Republic of Korea

⁶ Samsung Advanced Institute of Technology, Suwon 16678, Korea

⁷ Ewha Womans University, Seoul 03760, Republic of Korea

⁸ Department of Nano Science and Technology, Sungkyunkwan University, Suwon 16419, Republic of Korea

⁹ Department of Nano Engineering, Sungkyunkwan University, Suwon 16419, Republic of Korea

E-mail: jeongah.seo@mat.ethz.ch

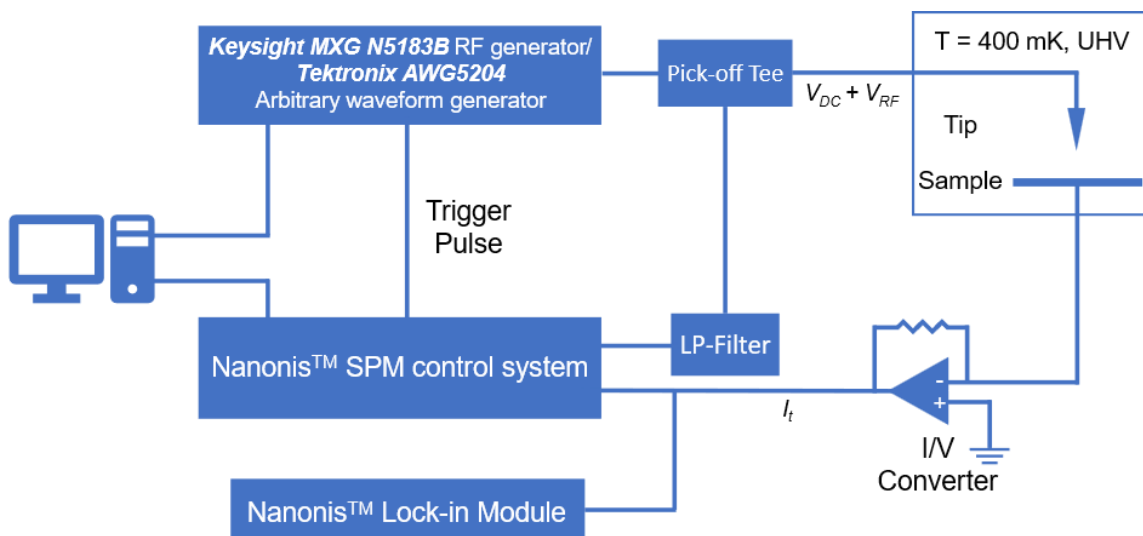
Nitrogen-doped graphene has sparked great interest in graphene application due to its unique physical and chemical properties. Owing to its tuneable features which depend on the atomic structure of nitrogen defects, it is important to control the doping configurations and understand the corresponding properties in order to utilize the potential of nitrogen-doped graphene in various applications. In this study, we investigated the nitrogen defects formed in graphene grown on Pt (111) surface using pyridine (C₅H₅N) as a nitrogen-containing precursor. Through simultaneous measurement of scanning tunneling microscopy (STM) and atomic force microscopy (AFM), we could compare the atomic structures of defects with their electronic structures. Notably, we were able to clearly identify two different types of nitrogen defects: graphitic-N and pyridinic-N. Atomic resolution of AFM imaging confirmed the atomic arrangement of each defect, which was not clearly resolved in STM imaging. In addition, the results of theoretical calculation using density functional theory (DFT) were consistent with our experimental results and helped in identifying the defects. Moreover, we could observe the dissociated pyridine precursor prior to forming graphene, which provided insight into the growth mechanism and explained the density of nitrogen defects.

Enhancing ESR-STM Measurements with Python Instrument Control Library

S. Shan¹, C. Soulard¹, J. Schwenk¹, and H. Brune¹

¹ *Laboratory of Nanostructures at Surfaces (LNS), Institute of Physics (IPHYS), Ecole Polytechnique Fédérale de Lausanne (EPFL), Switzerland*

Modern scanning probe microscopy (SPM) measurements become more and more sophisticated and involve configuring various instruments. For example, using a scanning tunneling microscope (STM) to do electron spin resonance (ESR) or stochastic resonance (SR) measurements typically demands configuring numerous software and instruments, such as Nanonis Software, radio frequency (RF) signal generators, arbitrary waveform generators, magnet PSU, etc. We present a Python library specifically designed to incorporate the software and instruments used in SPM measurements. In Nanonis SPM V5 Software, low-latency communication using the Transmission Control Protocol (TCP) can be established through the TCP server ports provided within the software. Similarly, other instruments, e.g., RF generator and AWG, allow for operations over TCP with Standard Commands for Programmable Instruments (SCPI). By utilizing this library, we are able to customize, streamline, and automate a wide range of measurements in a way that would be difficult to achieve via manual control or a graphical user interface. These include, but are not limited to, transfer function measurement, frequency sweeps, scanning tunneling spectroscopy, scan image acquisition, as well as operations like tip shaping and atom pickup. The integration of the Python library significantly improves efficiency, enhances productivity, and mitigates user errors.



References

- [1] Nanonis TCP protocol documentation, R12202, SPECS, (2022).

Coupling Single-Cycle Terahertz Fields into a Scanning Tunneling Microscope: Characterization and Time-Resolved Measurements

Junyoung Sim, Vibhuti Rai, Carlos Williams, Sergey Trishin, Nils Bogdanoff, Tom Seifert, Tobias Kampfrath, Christian Lotze, Katharina Franke

Freie Universität Berlin, Department of Physics, Arnimallee 14, 14195 Berlin, Germany

Local variations of electronic states and their dynamics play key roles to understand low dimensional system. Scanning tunneling microscopy (STM) is capable of accessing atomic-scale structures and electronic excitations on surfaces. However, there has been an instrumental limit of temporal resolution of excitation and relaxation processes due to stray capacitances of the wires and limited bandwidths of the amplifiers. To overcome this limitation, Cocker et al. coupled ultrashort Terahertz (THz) pulses into a conventional STM and implemented a pump-probe detection scheme [1].

In this work, we follow a similar approach by setting up a THz source and coupling it to a 5K-STM. Here, we provide a preliminary characterization of our set up with a few benchmarking experiments. We measure the transient voltage of the near-field THz pulse that modulates the photocurrent between STM tip and Ag(111) surface, which was pumped by a preceding optical pulse. We also probe the transient voltage in the regime of field emission resonance.

We then performed infrared-pump THz-probe measurement to investigate the dynamics of excitations on the semiconducting transition metal dichalcogenide MoTe₂. As we sweep the temporal delay between pump and probe pulses and vary their power, we measure the lifetime of excitation processes across the direct and indirect bandgap of the material [2].

Our preliminary results using THz-STM show that the technique provides rich possibilities to study dynamics in nanoscale structures.

References

- [1] Cocker *et al.* Nature Photonics 15, 558-569 (2021)
- [2] Sankar *et al.* Chem. Mater., 29, 2, 699–707 (2017)

STM method to measure the lifetime of a single nuclear spin

Evert. W. Stolte¹, Lukas. M. Veldman², Philip Willke³, Sander Otte¹

¹*Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology; 2628 CJ Delft, The Netherlands.*

²*Institute for Functional Matter and Quantum Technologies, University of Stuttgart, 70569 Stuttgart, Germany.*

³*Physikalisches Institut, Karlsruhe Institute of Technology; 76131 Karlsruhe, Germany.*

Electron Spin Resonance combined with Scanning Tunneling Microscopy (ESR-STM) has emerged as a promising platform for quantum coherent sensing at the nanoscale. However this application has been limited by the short coherence times of electron spins on metallic surfaces [1].

Nuclear spins on such surfaces, their state population accessible to ESR-STM already through the hyperfine interaction [2], might potentially have longer coherence time than electronic spins because they are protected from the two main sources of decoherence in ESR-STM: orbital electrons shield external magnetic field fluctuations from a vibrating STM tip and nuclear spins have no direct coupling to the electron bath of the metallic environment.

To uncover this potential, we investigate the relaxation time of a nuclear spin of ⁴⁷Ti on MgO/Ag. Recent research on this system found efficient nuclear spin pumping by electron scattering with the ⁴⁷Ti placed on the oxygen site of the MgO surface, polarising the nuclear spin to an excited state [3].

The nuclear spin relaxation time has thus far only been probed indirectly in experiments where the decoherence is dominated by an electron coupled to the nuclear spin.[2] We present a method to measure independently the nuclear spin relaxation in the time domain. As a work in progress, this method has resulted in a probable experimental lower bound for the relaxation time of 50 μ s.

References

- [1] Y. Chen, Y. Bae, A.J. Heinrich, *Adv. Mater.* **35**, 2107534 (2023),
- [2] P. Willke *et al.*, *Science* **362**, 336-339 (2018)
- [3] L.M. Veldman *et al.*, *Arxiv:2309.03749* (2023)

Local Electronic Structure of h-BN on Pt(110)

M. Thaler¹, F. Mittendorfer², E. Bertel¹ and L. Patera¹

¹*Institute of Physical Chemistry, Innsbruck, Austria*

²*Institute of Applied Physics and Center for
Computational Materials Science, Vienna, Austria*

The growth of large-scale single-domain hexagonal boron nitride (h-BN) monolayer has been recently achieved on the Pt(110) surface[1][2]. The resemblance between the reported band dispersion of h-BN/Pt(110) and the one calculated for a freestanding h-BN monolayer indicates mostly van der Waals interaction between boron nitride and Pt. However, a more localized bonding contribution is suggested by the fact that the substrate adapts to the h-BN adlayer to form a (1×n)-missing-row-reconstructed Pt(110) surface.

Here we exploited low-temperature scanning tunnelling spectroscopy (STS) and noncontact atomic force microscopy (AFM) to probe the morphology and local electronic structure of h-BN/Pt(110). The moiré pattern arising from the mismatch with the substrate is observed to induce a modulation of the conduction band onset of h-BN and a shift of a Pt-related state, implying varying interaction energies. AFM force spectroscopy reveals a topographic corrugation between the two distinct moiré regions. In agreement with density functional theory, the local density of states near the Fermi energy indicates a localized bonding between h-BN and Pt.

References

- [1] D. Steiner et al., ACS Nano **13**, 7083-7090 (2019)
- [2] M. Thaler, Phys. Rev. Research **2**, 043156 (2020)

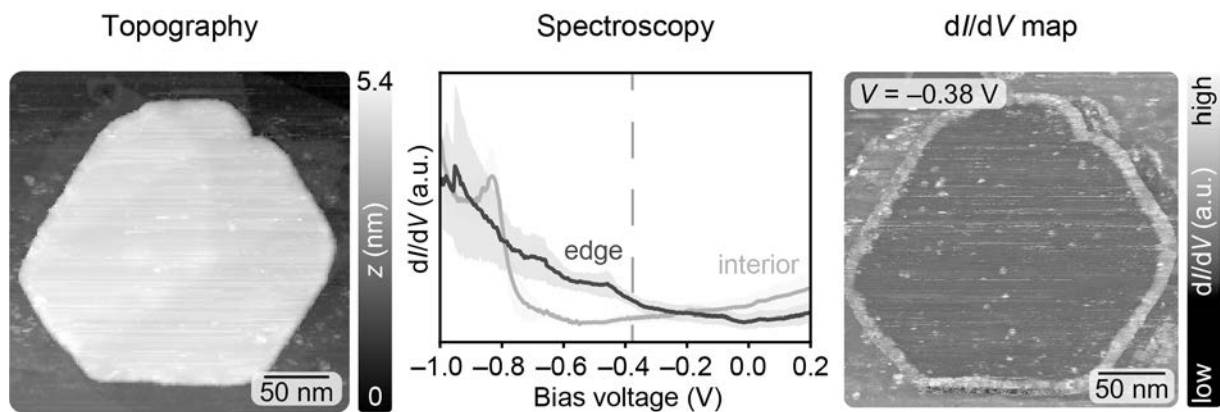
On the Edge State of Finite 2D Bi₂Se₃ Crystals

A. Vlasblom¹, J.R. Moes¹, J.F. Vliem¹, T.C. Wigmans¹, E.F. van Brenk¹, D. Vanmaekelbergh¹, I. Swart¹

¹Utrecht University, Utrecht, The Netherlands
E-mail: a.vlasblom@uu.nl

Topological insulators in two or three dimensions exhibit an insulating bulk, but possess gapless conducting edge or surface states that are protected by time-reversal symmetry.^[1] One property of the surface/edge state is spin momentum locking, which results in two types of surface currents, each with opposite spin (up or down) and opposite momentum (direction). In the 2D regime, top and bottom states hybridize and the interior becomes insulating. The two remaining types of edge currents are fully protected from backscattering by non-magnetic impurities.

Here, we study thin (3–6 quintuple layers) finite sized Bi₂Se₃ crystals, prepared via wet-chemical synthesis^[2], using scanning tunneling microscopy and spectroscopy. Measurements are performed on Bi₂Se₃ nanoplatelets with varying thickness to study the transition between 3D and 2D topological insulators. Additionally, the robustness of the edge states is investigated with respect to structural defects and deposition of magnetic impurities. These measurements provide new insights into the topological nature of 2D topological insulators.



References

- [1] Mazumder, K., Journal of Alloys and Compounds, **888**, 161492, (2021).
- [2] Zhang, J., Nano letters, **11(6)**, 2407-2414, (2011).

Phonon gap supported tunneling and Faraday screening through graphene

Tobias Wichmann^{1,2,3}, Keda Jin^{1,2,4}, Jose Martinez-Castro^{1,2,4}, Honey Boban⁵, Tom G. Saunderson^{6,7}, Yuriy Mokrousov^{6,7}, Lukasz Plucinski⁵, Markus Ternes^{1,2,4}, F. Stefan Tautz^{1,2,3} and Felix Lüpke^{1,2}

¹*Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich*

²*Jülich Aachen Research Alliance, Fundamentals of Future Information Technology*

³*Institut für Experimentalphysik IV A, RWTH Aachen*

⁴*Institut für Experimentalphysik II B, RWTH Aachen*

⁵*Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich*

⁶*Institute of Physics, Johannes Gutenberg University Mainz*

⁷*Peter Grünberg Institut (PGI-1) and Institute for Advanced Simulation (IAS)*

Encapsulation of van der Waals materials has proven a vital technique to protect them from degradation and contamination. Usually, metallic encapsulation layers mask the properties of the underlying material when studied in scanning tunneling microscopy. Utilizing the inelastic tunneling phonon gap of graphene, however, enables the unfettered investigation of low energy phenomena (e.g. Kondo effect, Majoranas, etc.) by scanning tunneling spectroscopy, while maintaining the advantages of encapsulated samples. Furthermore, we find that the conductive nature of the graphene encapsulation layer screens the sample from tip-induced electric fields, exemplified by our low-temperature STM examination of encapsulated Fe₃GeTe₂.