O 59: Spins on Surfaces at the Atomic Scale I

Time: Wednesday 10:30–13:00 Location: H6

O 59.1 Wed 10:30 H6

Quantum spin engineering in bottom-up assembled molecular nanostructures — •Taner Esat[1,2], Dmitriy Borodin[3,4], Jeongmin Oh[1,2], Andreas Heinrich[3,4], Stefan Tautz[1,2,5], Yujeong Bae[3,4], and Ruslan Temirov[1,2,6] — [1]Peter Grünberg Institute (PGI-3), Forschungszentrum Jülich, Germany — [2]Jülich Aachen Research Alliance, Germany — [3]Center for Quantum Nanoscience, Institute for Basic Science, South Korea — [4]Ewha Womans University, South Korea — [5]RWTH Aachen University, Germany — [6]University of Cologne, Germany

Scanning tunneling microscopy (STM) is a powerful technique for fabricating and studying artificial nanostructures with purpose-engineered quantum states. Using the manipulation capabilities of the STM, we place aromatic molecules in an upright geometry on a pedestal of two transition metal atoms on the surface [1] and on the STM tip. These nanostructures carry electron spins that are well decoupled from the metallic substrate [2]. Based on this, we fabricate a single-molecule quantum sensor at the apex of the STM tip and address it by electron spin resonance. We use this sensor to measure the magnetic and electric dipole fields emanating from a single atom with sub-angstrom spatial resolution [3]. Finally, we show that varying the transition metal atoms in the pedestal alters the spin state of the molecular nanostructures, leading to exceptionally long spin lifetimes of up to several minutes. [1] Esat et al., Nature 558, 573 (2018) [2] Esat et al., Phys. Rev. Research 5, 033200 (2023) [3] Esat et al., Nat. Nanotechnol. 19, 1466 (2024)

O 59.2 Wed 10:45 H6

Tuning the Kondo temperature of Porphyrin molecules via adsorption configurations. — •Xiangzhi Meng[1], Jenny Möller[2], Rodrigo Menchón[3], Alexander Weismann[1], Daniel Sánchez-Portal[3], Aran Garcia-Lekue[3], Rainer Herges[2], and Richard Berndt[1] — [1]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität, 24098 Kiel, Germany — [2]Otto-Diels-Institut für Organische Chemie, Christian-Albrechts-Universität, 24098 Kiel, Germany — [3]Donostia International Physics Center (DIPC), 20018 Donostia-San Sebastián, Spain

Magnetic molecules may serve as building blocks for spintronic devices at the ultimate limit of miniaturization. This sort of application requires an understanding of their spin properties. An important example is the Kondo effect that originates from the exchange interaction between a localized magnetic moment and the conduction electrons of the host metal. The interaction strength is reflected by a characteristic temperature, the Kondo temperature TK. In this work, we will show the Kondo effect of cobalt(II)-5,15-bis(4'-bromophenyl)-10,20-bis(4'-iodophenyl)porphyrin (CoTPPBr2I2) molecules on an Au(111) surface with a low-temperature STM. In comparison with the molecules reported before, the Kondo temperature of CoTPPBr2I2 can be tuned over a much broader range (8 K to 250 K) by switching their configurations. Additionally, we show that surface reconstruction plays a crucial role in modulating the molecular Kondo effect.

O 59.3 Wed 11:00 H6

Fock-Darwin states in tunable artificial atoms with high spin-orbit coupling — •Julian H. Strik, Hermann Osterhage, Kira Junghans, Niels P.E. van Mullekom, Emil Sierda, Anna M.H. Krieg, Danis Badrtdinov, Daniel Wegner, Mikhail I. Katsnelson, Malte Rösner, and Alexander A. Khajetoorians — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Quantum simulators are a pathway to study novel physical phenomena which are difficult to predict or observe in synthesized materials. To date, there is still a lack of viable platforms for quantum simulation to study confined electrons in strong magnetic fields, namely in the limit that the magnetic length is on the order of the Bohr radius. For typical crystals, this corresponds to field strengths that are unattainable in a laboratory.

In this talk I will discuss the magnetic response of atomic-scale quantum dots based on individual Cs atoms on the semiconducting surface of InSb [1]. Using low-temperature scanning tunnelling microscopy and spectroscopy, we pattern quantum dots of various sizes and geometries, by sculpting the potential of the underlying 2DEG. We then study the LDOS of these various structures to an external magnetic field. We relate this to the Fock-Darwin model of atomic states [2]. We review the experimental results in this context and discuss the role of structure size, symmetry and spin-orbit coupling.

[1] E. Sierda, et al, Science 380, 1048 (2023).
[2] L. P. Kouwenhoven et al., Rep. Prog. Phys. 64, 6 (2001).

O 59.4 Wed 11:15 H6

Nickelocene-functionalized tips as a molecular spin sensor — •Diego Soler-Polo[1], Ana Barragán[2], Andrés Pinar Solé[1], Manish Kumar[1], Oleksandr Stetsovych[1], Ben Lowe[1], Elena Pérez-Elvira[2], Koen Lauwaet[2], Pavel Jelínek[2], and José Ignacio Urgel[2] — [1]Institute of Physics of the Czech Academy of Sciences, Prague, Czech Republic — [2]IMDEA Nanoscience, Cantoblanco, Madrid, Spain

Polyradicals nanographenes with low-lying spin excitations have almost degenerate states with different spins. Even Many-Body methods such as CASSCF and NEVPT2 might not provide enough accuracy to determine the spin of the real ground state. Here, we demonstrate that scanning probe microscopy with a nickelocene-functionalized tip can distinguish between nearly degenerate spin states of single molecular π-magnets. The nickelocene molecule has a spin 1 and a small magnetic anisotropy of 4 meV that interacts with the molecular spins of the sample. The spectroscopic patterns as we the tip is approached can be related to the number of radicals and their couplings. Such patterns are well reproduced by simple spin models and the corresponding simulated inelastic current. First, the molecular radicals are fitted to a spin hamiltonian, which is then coupled to the S=1 modelling the nickelocene. A cotunneling formalism adapted to spin hamiltonians provides the dIdV maps comparable with the experimental measurements, thus revealing the spin of the sampled system.

O 59.5 Wed 11:30 H6

Vibrational excitations in magnetic triangular nanographenes — Nils Krane[1], •Elia Turco[1], Annika Bernhardt[2], Michal Juríček[2], Roman Fasel[1,3], and Pascal Ruffieux[1] — [1]Empa - nanotech@surfaces Laboratory, 8600 Dübendorf, Switzerland — [2]University of Zurich, Zurich, Switzerland — [3]University of Bern, Bern, Switzerland

Inelastic electron tunneling spectroscopy has become a powerful tool for the investigation of excited states in single molecules and other quantum dot systems. These excited states can be of various origins, notably from higher energy spin states but also from vibrational modes. Especially in the case of carbon-based molecules, featuring π-magnetism with strong exchange coupling, both excitation mechanisms can be found within one system. Here, we present a simple method to distinguish between these two mechanisms by spatial mapping of the excited states without any need for high magnetic fields. As a model system, we investigate the spin $S = 1/2$ phenalenyl radical on Au(111) and observe the featuring of two excited states via inelastic tun-neling spectroscopy. Comparison with DFT calculations proofs the vibrational origin of the observed inelastic features and allows us to assign them to distinct vibrational modes.

O 59.6 Wed 11:45 H6

Realization of separation of time scales in a heterogeneous atomic Boltzmann machine — •Kira Junghans, Hermann Osterhage, Werner M. J. van Weerdenburg, Niek M. M. Aarts, Anna M. H. Krieg, Tren Jacobs, Niels P. E. van Mullekom, Ruben Christianen, Eduardo J. Domínguez Vázquez, Hilbert J. Kappen, and Alexander A. Khajetoorians — Radboud University, Nijmegen, The Netherlands

The Boltzmann machine (BM) describes an energy-based neural network formed by coupled, fluctuating Ising spins [1]. A material realization of the Boltzmann machine was constructed using the complex stochastic dynamics of coupled Co atoms on the surface of black phosphorus (BP) [1]. Neurons and synapses were realized by a separation of time scales exploiting the anisotropic electronic structure of BP [1]. Using the anisotropy of the substrate is a limitation in scaling this concept.

Here, we study the coupled dynamics of different types of atoms on BP, which exhibit orbital memory [2,3]. We study the influence of

the coupling on stochastic dynamics in heteroatomic dimer and trimer configurations with scanning tunneling microscopy (STM). Further, we discuss how the multi-well energy landscape can be influenced by an applied AC signal.

[1] B. Kiraly et al., Nat. Nanotechn. 16, 414 (2021).
[2] B. Kiraly et al., Nat. Comm. 9, 3904 (2018).
[3] B. Kiraly et al., Phys. Rev. Research 4, 033047 (2022).

O 59.7 Wed 12:00 H6

Spin-surface interactions of S=1/2 molecular magnets on superconductors — •Susanne Baumann[1], Lukas Arnhold[1], Nicolaj Betz[1,2], Matteo Briganti[3], Andrea Sorrentino[4], Giulia Serrano[4], Federico Totti[3], Roberta Sessoli[3], and Sebastian Loth[1,2] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Center for Integrated Quantum Science and Technology, Stuttgart, Germany — [3]Department of Chemistry 'Ugo Schiff', University of Florence, Italy — [4]Department of Industrial Engineering, University of Florence, Italy

The interaction between magnetic molecules and superconducting surfaces critically depends on the electronic properties of the surface and the molecules' binding configuration, which determines the wave function overlap between molecule and surface. Using scanning tunneling microscopy (STM), we study the organometallic molecule ((η8-cyclooctatetraene)(η5-cyclopentadienyl)titanium) (CpTicot) on two different superconducting surfaces. On lead (Pb) nanoislands on Si(111), CpTicot exhibits multiple binding orientations with varying surface coupling strengths, that can be strong enough to generate Yu-Shiba-Rusinov bound states within the superconducting gap. Conversely, on vanadium (V(100)), the molecules adsorb in a single orientation with minimal coupling to the superconductor. Their spin states remain largely decoupled from the substrate, preserving their S=1/2 properties. These findings offer valuable insights into chemical design principles for molecular qubits where individual adressability and decoupling from superconducting substrates is desired.

O 59.8 Wed 12:15 H6

Edge states in bottom-up designed spin chains on a superconducting Rashba surface alloy — •Harim Jang, Khai That Ton, Lucas Schneider, Jens Wiebe, and Roland Wiesendanger — Department of Physics, University of Hamburg, Hamburg, Germany

The experimental quest for topological superconductors and emergent Majorana modes (MMs) at their edges have attracted significant interest recently in both directions of fundamental understanding of topology-driven quantum states and their potential applications in quantum computation leveraging the robustness by topological protection. A spin chain on a conventional superconductor in the presence of spin-orbit coupling is one of the promising platforms for realizing topologically non-trivial Yu-Shiba-Rusinov (YSR) bands, which can show signatures of MMs at the chain boundaries in the form of zero-energy states [1]. In this talk, we report on bottom-up designed Fe chains on the Rashba surface-alloy $BiAg_2$ grown on Ag(111)/Nb(110) and the detailed investigation of prominent states near the Fermi level at both chain ends, which are studied using scanning tunneling spectroscopy at sub-Kelvin temperatures. The atomically constructed Fe chains on the $BiAg_2$ alloy with proximity-induced superconductivity originating from the bulk Nb substrate show YSR bands with minigaps on the verge of our energy resolution inside the chain and edge states near the Fermi level at the chain's ends. The results are systematically compared to chains composed of other $3d$ transition metals, Mn and Co, on the same surface, revealing the absence of edge states for both cases. [1] S. Rachel and R. Wiesendanger, Phys. Rep. 1099, 1 (2025)

O 59.9 Wed 12:30 H6

Second harmonic driving of paramagnetic resonance of molecular spin through non-linear transport phenomena — •Stepan Kovarik[1], Richard Schlitz[1], Jose Reina-Gálvez[2,3], Aishwarya Vishwakarma[1], Dominic Ruckert[1], Nicolas Lorente[4,5], Christoph Wolf[2,3], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]ETH Zurich, Switzerland — [2]QNS, Seoul, Korea — [3]Ewha Woman University, Seoul, Korea — [4]CFM, Donostia-San Sebastian, Spain — [5]DIPC, Donostia-San Sebastian, Spain

The second harmonic excitation of electron paramagnetic resonance (EPR) originates from nonlinear processes. In this talk, I will demonstrate the presence of second and higher harmonic driving of a single spin in a pentacene molecule on 2ML MgO on Ag(001) when applying a radio-frequency electric field in an STM. Comparing our results to a theory considering EPR driven by a periodic modulation of the tunneling barrier [1] yields qualitative agreement, indicating the presence of higher harmonic driving. The key observation is that the barrier modulation can introduce nonlinearities to the electronic transport, leading to the excitation of magnetic resonance at the driving frequency [2] and at its multiples. Our work enhances the capabilities of EPR measurements in STM by introducing the frequency upconversion mechanism, showcasing the key role of electronic transport in driving the EPR. The presented findings apply also to other quantum transport systems, where the electrons are transported through discrete energy levels. References: [1]J. Reina-Gálvez et al., Phys. Rev. B 107, 235404 (2023)., [2]S. Kovarik et al., Science 384, 1368-1373 (2024).

O 59.10 Wed 12:45 H6

A quantum simulator to study electronic structure of matter in the Hofstadter limit — •Hermann Osterhage, Julian H. Strik, Kira Junghans, Niels P. E. van Mullekom, Anna M. H. Krieg, Emil Sierda, Danis Badrtdinov, Daniel Wegner, Mikhail I. Katsnelson, Malte Rösner, and Alexander A. Khajetoorians — Radboud University, Nijmegen, The Netherlands

The Hofstadter limit describes electronic structure in strong magnetic fields, where the magnetic length is on the order of the periodicity of the crystal. In this limit, the electronic structure shows self-similarity, namely fractal behavior. Experimentally, it is challenging to reach this limit for conventional crystals due to the high required field strengths. Therefore, one solution is to investigate structures with effectively larger periodicities. [1]. However, finding platforms that allow to study this limit with control over orbital and lattice symmetries is a current challenge.

Here, I will present a quantum simulator based on patterning Cs atoms on the surface of InSb(110) by scanning tunneling microscopy [2]. Cs atoms can be patterned into nanostructures that exhibit multi-orbital wavefunctions reminiscent of an artificial atom, and can be coupled to each other. We detail how the electronic spectrum evolves in magnetic field, and discuss the role of finite size effects and temperature, as well as link this to the expected spectra in the Hofstadter limit.

[1] R. Krishna Kumar et al., Science 357, 181 (2017).
[2] E. Sierda, et al, Science 380, 1048 (2023).