O 17: Spins on surfaces at the atomic scale – Poster

Time: Monday 18:00–20:00 Location: P2

O 17.1 Mon 18:00 P2

Conformational Control of Spin Lifetimes of a Single Molecule Magnet on a Metal Surface — •Sufyan Shehada[1], Corina Urdaniz[2], Manuel dos Santos Dias[3], Christoph Wolf[2], Dmitriy Borodin[2], Andreas Heinrich[2], Ruslan Temirov[1], Jeongmin Oh[1], Yujeong Bae[2], F. Stefan Tautz[1], Taner Esat[1], and Samir Lounis[4] — [1]Peter Grünberg Institute (PGI-3), Forschungszentrum Jülich, Germany — [2]IBS Center for Quantum Nanoscience (QNS), South Korea — [3]Scientific Computing Department, STFC Daresbury Laboratory, United Kingdom — [4]Institute of Physics, Martin-Luther-University Halle-Wittenberg, Germany

We present a first-principles study of a spin-engineered molecular nanostructure: a PTCDA molecule coordinated to two Fe atoms on an Ag(111) surface. This work complements recent STM-based quantum sensing experiments, where upright molecules were assembled on metallic pedestals to enable precise spin manipulation and nanoscale magnetic field detection using electron spin resonance [1-3]. Using density functional theory (DFT), we investigate how adsorption geometry influences exchange interactions and magnetic anisotropy energy in both standing and lying conformations. Incorporating DFT-derived parameters and experimental input, we developed a spin model to simulate spin relaxation dynamics [4-6]. This reveals how conformational control can regulate spin lifetimes at the single-molecule level.

[1] Esat et al., Nature '18. [2] Esat et al., PRR '23.[3] Esat et al., Nat. Nanotechnol. '24. [4] Khajetoorians et al., Science '13.[5] Delgado et al., Surf. Sci. '14.[6] Hermenau et al., Nat. Commun. '17.

O 17.2 Mon 18:00 P2

Towards faster dynamics of magnons in atomic spin chains — Christina Mier[1], Alexandra Meerovici Goryn[1], •Michael Schelchshorn[1], Lukas Veldman[2], and Sander Otte[1] — [1]Delft University of Technology, Delft, The Netherlands. — [2]University of Stuttgart, Stuttgart, Germany.

The coherent free evolution of coupled atomic spins [1] as well as electron and nuclear spins within a single atom [2] has previously been demonstrated using ESR-STM. Extending this approach to atomically assembled spin chains, provides a tunable platform for investigating magnon transport and interactions; by exploiting the spectral resolution of ESR-STM, we achieve controlled excitation and tuning of coherent spin-wave modes while pump-probe techniques allow us to directly map their dynamics on a nanosecond time scale. In particular, by engineering stronger exchange coupling both within the chain itself and to the readout atom, we observe many more oscillations within the coherence times, demonstrating dynamical timescales significantly faster than those reported by Veldman et al. [1]. This work addresses the coherent magnon dynamics within precisely engineered atomic spin chains and paves the way for probing more complex collective excitations in larger spin systems.

[1] Veldman, L.M., et al., Science, 2021. 372(6545): p. 964-968.

[2] Veldman, L.M., et al., Nature Communications, 2024. 15(1): p. 7951.

O 17.3 Mon 18:00 P2

Development of Time-Resolved Pump-probe SP-STS for Ultrafast Spin Dynamics in SAM — •Yannick Noettger[1] and Artem Odobesko[2] — [1]Wuerzburg University — [2]Wuerzburg University

Achieving coherent control of Single Atom Magnets (SAMs) is necessary for their use in quantum technologies and requires fundamental understanding of their spin dynamics on the atomic scale. Scanning tunneling microscopy (STM) provides a unique platform for probing and manipulating SAMs with atomic precision. One major challenge is minimizing coupling of the SAM to the surrounding as substrate-mediated relaxation strongly limits coherence and lifetime. Usually this is done by placing the SAM on a thin insulating layer on a metal substrate. We used an extended approach by combining the decoupling layer with a superconducting substrate, potentially enabling enhanced lifetimes due to the gap in the SC density of states. We investigate NaCl/Cu(111)/NbOx heterostructures grown by e-beam evaporation, which exhibit substrate induced superconductivity within the Cu(111)-layer up to a film thickness of 3ML. NaCl deposition on Cu(111)/NbOx was achieved, however showing very different growth characteristics than on bare Cu(111). Furthermore we implemented an all-electrical pump-probe spectroscopy scheme into our existing STM setup demonstrating functionalization and precise synchronization with the Lock-In Amplifier, although pump-probe measurements on single Fe atoms on Cu2N/Cu(001) did not yield any measurable lifetimes, highlighting limitations of our current STM setup.

O 17.4 Mon 18:00 P2

Structural characterization of $Sc_3N@C_{80}$ on Ag(111) with scanning probe microscopy — •Caroline Hommel[1,2], Seong-Hyun Hong[1,4], Andrés Pinar Solé[1,2], Merve Ercelik[1,3], Robert Ranecki[1,2], Shinjae Nam[1,2], Dmitriy Borodin[1,2], Andreas Heinrich[1,3], and Lukas Spree[1,2] — [1]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), Seoul 03760, Republic of Korea — [2]Ewha Womans University, Seoul 03760, Republic of Korea — [3]Department of Physics, Ewha Womans University, Seoul 03760, Republic of Korea — [4]Department of Physics, Korea University, Seoul 03760, Republic of Korea

Endohedral fullerenes are a fascinating class of compounds, that consist of a number of atoms trapped inside a fullerene cage structure. Among them, the Nitride Cluster Fullerenes (NCF) stand out for their stability and relatively high production yield. $Sc_3N@I_h$-C_{80} is the most abundant species in this class. Its electronic structure and the relatively free rotation of the internal cluster make it a promising candidate for molecular electronics.

In the presented work, we use STM and AFM with a CO functionalized tip to characterize individual $Sc_3N@I_h$-C_{80} molecules on Ag(111). This allows us to resolve their adsorption configuration and study the interaction of cage, cluster, and substrate in highest detail.

Studying $Sc_3N@I_h$-C_{80} at the single molecule level provides direct access to its geometric and electronic characteristics, which are often hidden in bulk measurements.

O 17.5 Mon 18:00 P2

Study of correlated spin interactions in bottom-up assembled molecular structures — •Lars Pütz[1,2,3], Daria Sostina[1,2,3], Stefan Tautz[1,3,4], and Markus Ternes[1,2,3] — [1]Peter Grünberg Institut (PGI-3); Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Experimentalphysik II B, RWTH Aachen University, 52072 Aachen, Germany — [3]Jülich Aachen Research Alliance (JARA), Fundamenatls of Future Information Technology, 52425 Jülich Germany — [4]Experimentalphysik IV A, RWTH Aachen University, 52072 Aachen, Germany

Correlated physics hosts phenomena such as unconventional superconductivity and exotic magnetic phases, yet their theoretical description is often challenging. To provide a starting point, we investigate finite correlated systems. We study NTCDA molecules deposited on an Ag(111) surface using a low-temperature scanning tunneling microscope. These molecules can receive an additional electron from the surface, which turns each of them into a spin-1/2 system. Normally, this spin is screened by the substrate through the Kondo effect. By using molecular manipulation techniques, we built small molecular structures in which the spin-exchange interaction between two neighboring molecular spins overcomes the screening from the surface. In these structures, we observe singlet-triplet excitations. Moreover, these excitations exhibit a spatially varying bias asymmetry in scanning tunneling spectroscopy, reflecting the phase pattern of the triplet wave function and thus providing experimental access to it.

O 17.6 Mon 18:00 P2

Spectra and competing correlations in coupled magnetic impurities on superconducting substrates — •Carlos Quesada Pérez and David Jacob — Departamento de Física, Universidad de Alicante, Spain

The study of magnetic impurities in superconductors is a fundamental challenge in condensed matter physics, crucial for understanding phenomena like in-gap states such as Yu-Shiba-Rusinov (YSR) states. Systems with multiple impurities can potentially host exotic ground states relevant for topological quantum applications. Accurate numerical methods like the numerical renormalization group are computationally too demanding for solving more than two impurity levels. Recently, an efficient impurity solver was proposed, based on the exact

diagonalization of a single impurity connected to discretized superconducting reservoirs [1]. In this work, we extend this model to the case of two magnetic impurities coupled to BCS superconductors. By exact diagonalization of the resulting tight-binding Hamiltonian, we study the competition between the impurity-superconductor coupling and the impurity-impurity exchange to form a singlet with the bath or a singlet between the impurities themselves. The results reveal the evolution of YSR states and spin correlations, showing transitions between distinct quantum phases driven by this competition.

References:

[1] Phys. Rev. B **108**, L220506 (2023)

O 17.7 Mon 18:00 P2

Atomic-Scale Engineering of Spin Defects in Monolayer MoS_2 — •Johannes Schwenk[1], Wantong Huang[1], Kwan Ho Au-Yeung[1], Johanna Matusche[1], Paul Greule[1], Jonas Arnold[1], Lovis Hardeweg[1], Máté Stark[1], Luise Renz[1], Affan Safeer[2], Daniel Jansen[2], Jeison Fischer[2], Thomas Michely[2], Wolfgang Wernsdorfer[1], Christoph Sürgers[1], Wouter Jolie[2], and Philip Willke[1] — [1]Physikalisches Institut (PHI), Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany — [2]II. Physikalisches Institut, Universität zu Köln, Köln, Germany

Solid-state spin defects have become a promising platform for quantum technologies, including quantum sensing, communication, and computing. However, reliably creating and addressing such defects individually remains a major challenge. Here, we create distinct defect types in monolayer MoS_2 using the tip of a scanning tunnelling microscope, i.e. sulphur vacancies and substitutional impurities. Moreover, we characterize their spin properties using electron spin resonance scanning tunnelling microscopy (ESR-STM) and demonstrate coherent control of the spin defects. By constructing dimers of defects, we show that their spins can be coupled via magnetic exchange, which provides a path to realize larger spin structures. These results open a new avenue to design and engineer quantum states at the atomic scale for applications in quantum sensing and simulations [Nat. Nanotech.19,1782-1788 (2024)].

O 17.8 Mon 18:00 P2

Switching properties of a spin-crossover complex on Cu(111) analyzed from time traces of the tunneling current — •Katharina Biel[1], Jasmeen Jasmeen[1], Sujoy Karan[1], Karl Ridier[2], Gael Reecht[1], and Manuel Gruber[1] — [1]Faculty of Physics and CENIDE, University of Duisburg-Essen, Germany — [2]LCC, CNRS and Université de Toulouse, France

Spin-crossover molecules can be switched between two states associated with different spin, a low spin (LS) and a high spin (HS) state. The switching between these two states can be induced by tunneling electrons, among other triggers, which offer the possibility to use a scanning tunneling microscope (STM) to investigate the switching dynamics at the single molecular level.

Here we investigate the spin-crossover molecule [Fe(HB(1,2,4-trazolyl)$_3$)$_2$], which forms self-assembled islands on Cu(111) and exhibits efficient switching under electron injection. The switching is monitored via time traces of the tunneling current, exhibiting two levels corresponding to the different spin states. Statistical analysis of these traces provides a detailed view on the switching properties. In particular we studied the influence of the applied voltage and the tunneling current on the switching rate. The influence of the applied voltage and the tunneling current on the switching yield is studied in details.

O 17.9 Mon 18:00 P2

Spin-Electric Control of Individual Spins on Surfaces — Paul Greule[1], Wantong Huang[1], Máté Stark[1], Kwan Ho Au-Yeung[1], Johannes Schwenk[1], Jose Reina-Gálvez[2], Christoph Sürgers[1], Wolfgang Wernsdorfer[1], Christoph Wolf[3], and •Philip Willke[1] — [1]Physikalisches Institut (PHI), Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany — [2]Department of Physics, University of Konstanz, Konstanz, Germany — [3]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic of Korea.

Individual magnetic molecules are promising building blocks for quantum technologies because of their chemical tunability, nanoscale dimensions, and ability to self-assemble into ordered arrays. In this work, we present spin-electric coupling (SEC) for two molecular spin systems, iron phthalocyanine (FePc) and Fe-FePc complexes, adsorbed on a surface. We use electron spin resonance combined with scanning tunnelling microscopy (ESR-STM) to locally address them with the STM tip and electrically tune them using the applied bias voltage. These measurements reveal a pronounced nonlinear voltage dependence of the resonance frequency, linked to the energic onset of other molecular orbitals. We attribute this effect to a transportmediated exchange field from the magnetic tip, providing a large, highly localized, and broadly applicable SEC mechanism. Finally, we demonstrate that the SEC enables all-electrical coherent spin control: In Rabi oscillation measurements of both single and coupled Fe-FePc complexes we show that the spin dynamics can be tuned.

O 17.10 Mon 18:00 P2

Defluorination of F_{16}Cu-Phthalocyanine on Superconducting Pb(100) — •Julian Skolaut, Biswajit Pabi, Alexander Weismann, and Richard Berndt — Institute for Experimental and Applied Physics, CAU Kiel, Germany

F_{16}Cu-phthalocyanine was investigated on a superconducting Pb(100) surface using low-temperature scanning tunneling microscopy. We achieved controllable defluorination of selected lobes. A single defluorinated lobe serves as a pivot point for rotation toward eight different azimuthal orientations. Defluorination of more than one lobe eliminates the ability to rotate. The energy of the lowest unoccupied molecular orbital depends on the degree of defluorination and is lowest when two lobes have been defluorinated. We also observed Yu-Shiba-Rusinov states, which exhibit shifts depending on the adsorption configuration and azimuthal orientation.

O 17.11 Mon 18:00 P2

Investigations of FePc molecular spins on a superconducting substrate — •Luise Renz[1], Máté Stark[1], Jonas Arnold[1], Johannes Schwenk[1], Christoph Sürgers[1], Wolfgang Wernsdorfer[1], and Philip Willke[1,2] — [1]Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Center for Integrated Quantum Science and Technology (IQST), Karlsruhe Institute of Technology, Karlsruhe, Germany

Electron spin resonance (ESR) measurements performed with a scanning tunneling microscope (STM) has emerged as an excellent technique to probe quantum spin systems at the nanoscale. However, the spin lifetimes of surface spins are limited by scattering with substrate electrons[1]. This can be prevented by employing thin insulating layers[1]. Alternatively, superconducting substrates can be used for decoupling[2].

Here, we present results on FePc molecules on a superconducting substrate. First, we characterize the FePc molecules on the new substrate by investigating the Zeeman splitting and the molecular orbitals. Our findings show that FePc resembles a spin-1/2 system, and that the molecular orbitals exhibit a similar pattern as was previously reported on MgO/Ag(001)[3]. Additionally, we perform all electric pump-probe measurements on single FePc molecules. By adjusting the external magnetic field, we can switch between the normal conducting and superconducting state and investigate the influence of the superconductor on the T_1 time of the FePc molecules. [1] Nat. Phys. 13, 403 (2017) [2] Nat. Phys. 9, 765 (2013) [3] Nat Commun 16, 5208 (2025)

O 17.12 Mon 18:00 P2

Spin Screening in MoS_2 Mirror Twin Boundaries on Graphene/Ir(110) — •Kuber Vyas, Tfyeche Tounsi, Jeison Fischer, Affan Safeer, Thomas Michely, and Wouter Jolie — II. Physikalisches Institut, Universität zu Köln, Zülpicher Str. 77, 50937 Köln, Germany

Mirror twin boundaries (MTBs) are line defects that host confined states in the bandgap of monolayer MoS_2(1). The confined states closest to the Fermi energy splits into a singly and a doubly occupied level separated by the Coulomb repulsion U, forming a spin-1/2 system. The spin of the singly occupied state is then screened by the electron bath present in the substrate below, giving rise to a Kondo resonance pinned to the Fermi energy.

In this work, we investigate spin screening in MoS_2 MTBs grown on graphene/Ir(110) using low-temperature scanning tunnelling microscopy and spectroscopy. STS spectra recorded along the boundaries show two pronounced impurity peaks separated by the Coulomb energy U, together with a zero-bias feature which can be attributed to the Kondo resonance. MTBs on Gr/Ir(110) exhibit significantly broadened impurity-level widths, indicating strong hybridization γ with the electron bath provided by the graphene/Ir(110) substrate. This enhanced hybridization results in a Kondo temperature T_K comparable to the measurement temperature (7 K), enabling us to study the phase

transition between the Kondo singlet phase below and the magnetic phase above the Kondo temperature. [1] C. van Efferen et al, Nat. Phys. 20, 82-87 (2024)