O 45: Poster Spins on Surfaces at the Atomic Scale

Time: Tuesday 18:00–20:00 Location: P2

O 45.1 Tue 18:00 P2

Scanning Tunneling Microscopy and Spectroscopy of YbPc2 Molecules — •Jonas Arnold[1], Kwan Ho Au-Yeung[1], Wantong Huang[1], Paul Greule[1], Christoph Sürgers[1], Wofgang Wernsdorfer[1], Mario Ruben[2], and Philip Willke[1] — [1]Physikalisches Institut, Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Institute of Nanotechnology, Karlsruhe Institute of Technology, Karlsruhe, Germany

Individual molecules constitute excellent building blocks for quantum technologies thanks to their small size, reproducibility and the benefit of self assembly. A promising class are rare-earth bis-phythalocyanine complexes [1]. In this investigation, YbPc2 molecules are studied using scanning tunneling microscopy (STM) and spectroscopy (STS) to identify potential indicators of a magnetic signature. In the gas phase, the YbPc2 molecule is expected to exhibit a radical spin localized at its ligands as well as an f-shell electron spin S = 1/2 and a nuclear spin (I = 1/2 and 5/2) for certain isotopes. Thus, this system is a potential candidate for a spin cascade [1]. We perform measurements on self-assembled multi-layer islands of YbPc2 on Ag(100) that reveal distinct orbital features which vary for the first, second and third molecular layer. Similarly, the orbital signatures change when introducing a thin dielectric decoupling layer of MgO between the molecule islands and the electron bath. We discuss these results in the context of charge transfer between the molecular film and the underlying substrate [2]. [1] Wernsdorfer, W. et al., Advanced Materials 31, 1806687 (2019). [2] Hollerer, M. et al., ACS nano 11, 6252-6260 (2017).

O 45.2 Tue 18:00 P2

Magnetic adatom manipulation on monolayer transition metal dichalcogenides — •Daniel Jansen[1], Katharina Offermann[1], Tfyeche Tounsi[1], Affan Safeer[1], Jeison Fischer[1], Arkady Krasheninnikov[2], Nicolae Atodiresei[3], Thomas Michely[1], Hannu-Pekka Komsa[4], and Wouter Jolie[1] — [1]II. Physikalisches Institut, Universität zu Köln, Köln, Germany — [2]Institut für Ionenstrahlphysik und Materialforschung, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany — [3]Peter Grünberg Institut, Forschungszentrum Jülich, Jülich, Germany — [4]Faculty of Information Technology and Electrical Engineering, University of Oulu, Oulu, Finland

Two-dimensional materials are found to host a large variety of correlated phases. A promising approach towards understanding and controlling these phases is by means of atomic manipulation.
Here we compare two systems for atomic manipulation experiments: Fe on 1H-MoS_2 and Fe on 1H-TaS_2. We find that manipulation of Fe on 1H-MoS_2 results in point defects (sulfur vacancies) [1], which we investigate using scanning tunneling microscopy and spectroscopy. In contrast, we find that single Fe adatoms can be laterally moved on 1H-TaS_2, enabling construction of lattices consisting of tens of adatoms. We additionally report on the observation of two inequivalent adsorption sites for Fe, hollow and Ta-top sites, which manifest as differences in the adatom's apparent height.

[1] Jansen et al., Phys. Rev. B, **109**, 195430, 2024

O 45.3 Tue 18:00 P2

investigating the origins of spin-polarization in Au(111) : experiments vs theory — •Sourour Ayari[1], Laurent Nicolaï[1], Aki pulkkinen[1], Ridha Eddhib[1], Saleem Khan[1], Trung Phuc[1], Ján Minár[1], and Mauro Fanciulli[2,3] — [1]New Technologies - Research Centre, University of West Bohemia, 301 00 Pilsen, Czech Republic. — [2]CY Cergy Paris University, France — [3]CEA Paris Saclay, France

This study explores the origins of spin polarization in semi-infinite Au(111), examining whether the observed spin polarization arises primarily from the system's initial states or solely through the Photoemission process. To address this, we will integrate both experimental and theoretical results to provide an understanding of this spin-polarization origins, we calculate the electronic band structure with and without the influence of Mott-Scattering, while isolating additional factors, such as the Rashba effect[1]. On the theoretical side the calculations are performed using the SPRKKR method [2,3] which is based on DFT calculations, which will account for fundamental effects, while the additional one-step model will account for the photoemission process.

[1]E. E. Krasovskii and E. V. Chulkov Phys. Rev. B 83, 155401 (2011) [2] H. Ebert, D. Ködderitzsch and J. Minár, Rep. on Prog. in Phys. 74, 096501 (2011) [3] J. Braun et al., Phys. Rev. B 88, 205409 (2013)

O 45.4 Tue 18:00 P2

Coherent driving of interacting spins in single molecules — •Maria Steiner and Andrea Donarini — Institute for Theoretical Physics, University of Regensburg, Regensburg, Germany

The low energy spectrum of neutral closed shell molecules generically exhibits a set of singlet and triplet excited levels. The latter arises due to the exchange interaction between the unpaired electrons in singly occupied frontier orbitals. Moreover, the spatial anisotropy of these molecular states, combined with the spin-dipolar interaction, induces a zero-field splitting of the triplet. In recent AFM-ESR experiments on pentacene [1] Rabi oscillations between two of the split triplet states have been demonstrated. We now extend this concept to the full triplet space. By means of multiple frequency coherent driving, we investigate theoretically the rich dynamics of this molecular qutrit in search, for example, of the analogue of the dark states observed for Λ-systems in atomic physics.

[1] Sellies et al., Nature **624**, 64-68 (2023).

O 45.5 Tue 18:00 P2

A molecular spin on a scanning probe tip enables quantum sensing at the atomic scale — 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; Jülich, Germany — [2]Jülich Aachen Research Alliance (JARA), Fundamentals of Future Information Technology; Jülich, Germany — [3]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS); Seoul, South Korea — [4]Department of Physics, Ewha Womans University; Seoul, South Korea — [5]Experimentalphysik IV A, RWTH Aachen University; Aachen, Germany — [6]Faculty of Mathematics and Natural Sciences, Institute of Physics II, University of Cologne; Cologne, Germany

In this work, we fabricate a single-molecule quantum sensor on a scanning tunneling microscope (STM) tip by attaching Fe atoms and a PTCDA (3,4,9,10-perylenetetracarboxylic-dianhydride) molecule to the tip apex. The PTCDA molecule is a spin-1/2 system on the STM tip and serves as a two-level quantum system in a magnetic field. We probe this molecular spin system by electron spin resonance and achieve about 100 neV resolution in energy. The functionality of the quantum sensor we demonstrate by measuring the magnetic and electric dipole fields emanating from a single Fe atom and an Ag dimer on an Ag(111) surface with sub-angstrom spatial resolution [T. Esat, D. Borodin, J. Oh et al. Nat. Nanotechnol. 19, 1466 (2024)].

O 45.6 Tue 18:00 P2

Thermometry of a millikelvin scanning probe junction with spin-flip inelastic electron tunnelling spectroscopy — •Emilio Scontrino[1,3], Stefan Schulte[2,3], Taner Esat[3], Markus Ternes[1,3], Stefan Tautz[1,3], and Ruslan Temirov[2,3] — [1]RWTH Aachen — [2]Universität zu Köln — [3]Forschungszentrum Jülich, PGI-3, Germany

Organic molecules attached to the apex of a scanning probe tip have recently emerged as promising sensors for electric and magnetic fields at atomic scales [1, 2, 3]. Here, we functionalize the tip of a millikelvin scanning tunnelling microscope (STM) [4] with a 1,4,5,8-naphthalene tetracarboxylic dianhydride (NTCDA) molecule. We measure the inelastic spin-flip excitations of this molecular spin 1/2 system in an out-of-plane magnetic field of 7 T varying the temperature of the adiabatic demagnetization cryostat between 30 mK and 1.2 K. To perform thermometry of the STM junction we fit the temperature-dependent smearing of the spin-flip excitation spectra using the third-order perturbation theory in the Appelbaum-Anderson-Kondo framework [5].

[1] C. Wagner et al., Phys. Rev. Lett. 115, 026101 (2015) [2] B. Verlhac et al., Science 366 , 623 (2019) [3] T. Esat et al., Nat. Nano. 19, 1466 (2024) [4] T. Esat et al., Rev. of Sci. Instr. 92 (6) (2021) [5]

M. Ternes, New J. Phys. 17, 063016 (2015)

O 45.7 Tue 18:00 P2

Investigation of the Kondo effect in complex, artificially built, single molecule structures — •Lars Pütz[1], Daria Sostina[1], Taner Esat[1,2], Stefan Tautz[1,2,3], Ruslan Temirov[1,4], and Markus Ternes[1,2,5] — [1]Peter-Grünberg-Institute for Quantum Nanoscience, Research Center Jülich, 52425 Jülich, Germany — [2]Jülich Aachen Research Alliance, Fundamentals of Future Information Technology, 52425 Jülich, Germany — [3]Institute of Physics IV, RWTH Aachen University, 52074 Aachen, Germany — [4]Institute of Physics II, University of Cologne, 50937 Cologne, Germany — [5]Institute of Physics IIB, RWTH Aachen University, 52074 Aachen, Germany

In engineered spin systems, strongly correlated many-body phenomena such as nontrivial topology, Kondo screening and highly correlated long-range states can be constructed and studied by site-selective measurements with scanning tunneling microscope (STM) tips [C. Zhao et al., Nat. Nanotech. 2024]. Here we show how many-body phenomena evolve by controlled manipulation of NTCDA molecules on Ag(111) using the tip of an STM. Building small clusters of 2-5 molecules and tuning their spatial geometry reveals complexly patterned Kondo resonances and, in addition, an electric field-dependent strong increase in conductance, which we attribute to the collapse of Kondo correlations [S. Arabi et al., arXiv 2022]. Single NTCDA molecules on the other hand show no detectable spin signature, despite the clear formation of Kondo resonances in clusters where their high symmetry with respect to the surface is broken.

O 45.8 Tue 18:00 P2

Investigation of the Yu-Shiba-Rusinov states arising from single Fe atoms on superconducting $2H$-NbS_2 — •Margarete Huisinga, Werner M.J. van Weerdenburg, Lisa M. Rütten, and Katharina J. Franke — FachbereichPhysik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

Magnetic adatoms on a superconductor give rise to states within the superconducting gap due to the interaction of the unpaired spin(s) with the Cooper pairs of the substrate. These so-called Yu-Shiba-Rusinov (YSR) states are therefore isolated from the rest of the electronic structure of the system, which makes them interesting building blocks for artificial molecules or the design of band structures, if the YSR states of multiple atoms hybridize. The YSR states are especially long range on 2D materials, which enables hybridization over a large spacing. Previous experiments on the hybridization of YSR states performed on $NbSe_2$ were limited by a charge density wave [1], which is not present in $2H$-NbS_2.
Using scanning tunneling microscopy and spectroscopy, we investigate single Fe atoms on superconducting $2H$-NbS_2. We find that Fe adsorbs in two different adsorption sites on the substrate, each with a distinct set of YSR states. We map the spatial distribution of these YSR states to determine their symmetry and find long-ranged YSR features extending up to 4 nm. Additionally, we observe a tip-induced shift in the energy of the YSR states for one adsorption site species.

[1] Liebhaber et al. Nat Commun 13, 2160 (2022).

O 45.9 Tue 18:00 P2

Hybridization of Yu-Shiba-Rusinov states in magnetic clusters on Pb(111) — •Katharina Biel, Bharti Mahendru, Werner M. J. van Weerdenburg, Lisa M. Rütten, and Katharina J. Franke — Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

The unpaired spin of a magnetic impurity on a superconducting substrate interacts with the Cooper pairs via potential and exchange scattering. This leads to local in-gap bound states, so-called Yu-Shiba-Rusinov (YSR) states. The hybridization of close-by atoms can be described by linear combinations of the YSR states [1,2].

We investigated self-assembled manganese (Mn) and iron (Fe) clusters on superconducting lead (Pb(111)) by scanning tunneling microscopy and spectroscopy. While the topography essentially shows featureless protrusions, the differential conductance maps reflect characteristic shapes of the YSR states. We employ a phenomenological hybridization model [3] to simulate the observed shapes. In particular, we show atomic configurations which lead to chiral YSR patterns.

[1] Ruby, M., et al., PRL 120, 156803 (2018); [2] Amann, S., et al., PRB 108, 195403 (2023); [3] Rütten, L., et al., ACS Nano 18, 30798 (2024)

O 45.10 Tue 18:00 P2

Investigation of impurity states on the high-temperature superconductor $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ via scanning tunneling microscopy — •Melvin Grumser, Verena Caspari, Werner M. J. van Weerdenburg, and Katharina J. Franke — Freie Universität Berlin, Department of Physics, Arnimallee 14,14195 Berlin, Germany

The interaction of magnetic impurities and superconductors gives rise to impurity states in the superconducting gap, which have been widely studied for s-wave superconductors [1]. For d-wave superconductors however, only few experiments addressing the interplay of local spins and Cooper pairs, governed by an anisotropic order parameter and non-vanishing density of states (DOS) in the superconducting gap have been reported [2,3].
Here, we investigated the interaction between the high-temperature d-wave superconductor $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ and magnetic adatoms on the atomic scale using scanning tunneling microscopy. We present measurements conducted on scandium, iron and manganese adatoms, expected to display different amounts of in-gap states due to the different number of unpaired electrons. We find strong variations in the spectra that we ascribe to different adsorption sites and variations of the density of states across the substrate.

[1] W. Heinrich, I. Pascual, K. J. Franke, Progress in Surface Science 93 (2018)
[2]U. Erdenemunkh, M. C. Boyer, Physical review letters 117 (2016)
[3] J. Davis, Nature 411 (2001)

O 45.11 Tue 18:00 P2

Atomic-scale Josephson spectroscopy performed over magnetic clusters on Pb(111) — •Bharti Mahendru[1], Martina Trahms[1,2], Katharina Biel[1], Werner M. J. van Weerdenburg[1], Clemens B. Winkelmann[2], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany — [2]Université Grenoble Alpes, CNRS, Institut Neél, 25 Avenue des Martyrs, 38042 Grenoble, France

When two superconducting leads are brought in close proximity, they form a Josephson junction (JJ). In a JJ, quantum tunnelling of Cooper pairs occurs without an applied voltage between the two superconductors with a phase difference. To study the properties of Cooper-pair tunneling at the atomic scale, a scanning tunneling microscope is a powerful tool. A JJ forms by approaching the tip to the sample. Previously, it has been shown that a Josephson junction including a single magnetic adatom shows non-reciprocity in the retrapping current [1]. This expression of the Josephson-diode effect was ascribed to the broken particle-hole symmetry induced by the Yu-Shiba-Rusinov (YSR) states inside the superconducting energy gap. Here, we investigate the effect of hybridized YSR states of self-assembled Fe clusters on Pb(111). We find non-reciprocal retrapping currents that we correlate with the asymmetry in the YSR states and their spatial distribution.
[1] Martina Trahms et al., *Nature* 615, (2023)

O 45.12 Tue 18:00 P2

Driving nuclear spin transitions on a single atom using ESR-STM — •Hester Vennema, Cristina Mier Gonzalez, Evert Stolte, Rik Broekhoven, Jinwon Lee, and Sander Otte — Delft University of Technology, The Netherlands

The spin of a single nucleus is a prime candidate for quantum information applications due to its weak coupling to the environment and subsequently longer coherence times [1]. Using the high energy resolution of electron spin resonance (ESR) in combination with the high spatial resolution of scanning tunneling microscopy (STM), it is possible to measure the hyperfine interaction between the nucleus and an electron for single atoms on a surface [2].

A recent effort has successfully resolved the coherent dynamics of a hyperfine-driven interaction between nuclear and electron spin [3]. Moreover, the lifetime of the nuclear spin has been measured to be in the order of seconds [4].

In this study we use a double resonance measurement scheme to resolve nuclear spin transitions of a single 47Ti isotope with a spin of I=5/2. Additionally, we are able to selectively drive multiple transitions directly.

[1] Pla et al. (2013), Nature 496, 334-338
[2] Willke et al. (2018), Science 362, 336-339
[3] Veldman et al. (2024), Nat. Comm. 15, 7951
[4] E. Stolte et al. (2024), arXiv:2410.0870

O 45.13 Tue 18:00 P2

Nanoscale Control of Quantum States in Radical Molecules

on Superconducting Pb(111) — •Chao Li, Jung-Ching Liu, Outhmane Chahib, Thilo Glatzel, Rémy Pawlak, and Ernst Meyer — Department of Physics, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland.

Superconducting surfaces hosting magnetic impurities provide a promising framework for applications in quantum technology. Here, we demonstrate the manipulation of magnetic states in the radical molecule 4,5,9,10-tetrabromo-1,3,6,8-tetraazapyrene (TBTAP) deposited on a Pb(111) superconducting surface, utilizing low-temperature scanning tunneling microscopy. Tunneling spectroscopy reveals the presence of Yu-Shiba-Rusinov (YSR) states near the Fermi level for isolated TBTAP molecules. By varying the tip-molecule distance, we induce a quantum phase transition between singlet and doublet ground states. Furthermore, introducing a second TBTAP molecule enables tuning of the YSR state position through modifications in relative distance and orientation, with specific configurations leading to the splitting of YSR states. Extended molecular chains, up to pentamers, exhibit periodic patterns of charged and neutral molecules, where even-numbered chains form a distinct charged dimer structure at one terminus. This dimer position can be manipulated, allowing for information encoding within the chain.

O 45.14 Tue 18:00 P2

Exchange interactions of spin states in coupled triangular nanographenes — •Suyash Singh[1], Nils Krane[1], Elia Turco[1], Roman Fasel[1,2], and Pascal Ruffieux[1] — [1]Empa - Swiss Federal Laboratories for Materials Science and Technology, 8600 Dübendorf (Switzerland) — [2]Department of Chemistry and Biochemistry, University of Bern, 3012 Bern, Switzerland

Zigzag-edged triangular nanographenes (triangulenes) are promising realizations of spin qubits at a molecular level, with a total spin S scaling with the length of their edges. Chemical design and on-surface synthesis methods allow atomically precise engineering of carbon-based π–systems with strong spin-spin interactions, using triangulene units as building blocks. Exploiting the rules governing the exchange interactions, functional groups and spacer molecules, such as phenyl rings, can be used to tune the sign and magnitude of the exchange coupling between the triangulene units. We present the bottom-up fabrication of correlated spin platforms using the two smallest members of the triangulene family, the spin S=1/2 phenalenyl and S=1 [3]triangulene. We analyze the origin of spin-polarized states and spin excitations using a tight-binding and mean-field Hubbard Model for the π–orbital network. With an estimate of the couplings within the spin cluster, we describe it using an effective Heisenberg model. The spin excitations are then investigated experimentally using inelastic electron tunneling spectroscopy. Specifically, we summarize the results on dimers and trimers, where the exchange mechanisms can be investigated systematically by hydrogenation and selective re-activation of single spins.

O 45.15 Tue 18:00 P2

Properties of magnetic atoms on a hexagonal insulating lattice — •Johannes Schust, Henrik Lichtl, Julian Zeitler, Lukas Arnhold, Sebastian Loth, and Susanne Baumann — University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

The symmetry of the immediate surrounding of an atom heavily influences its static properties and also governs its dynamics. For the exploration of atomic-scale systems by scanning tunneling microscopy (STM), the choice of a suitable thin film is thus key. While most preceding works take advantage of two- or four-fold underlying symmetries, we focus on the hexagonal surface of zinc oxide (ZnO) monolayers. Hexagonal substrates bear the potential for exploring spin frustration effects on a surface, realized by the assembly of magnetic nanostructures built atom-by-atom. ZnO, as a wide bandgap semiconductor [1], is employed to mitigate electron scattering between the Ag(111) substrate and the adsorbed species on the sample. We develop a reliable method to prepare the aforementioned substrate and carry out comprehensive characterization of cobalt (Co) and manganese (Mn) atoms in the present environment. These efforts provide the foundation for investigating novel dynamical aspects of frustrated systems on the atomic scale.

[1] A. Shiotari et al. J. Phys. Chem. (2014)

O 45.16 Tue 18:00 P2

Multiplet calculations of magnetic atoms on a hexagonal insulator — •Julian Zeitler[1], Johannes Schust[1], Henrik Lichtl[1], Lukas Arnhold[1], Sebastian Loth[1,2], and Susanne Baumann[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Center for Integrated Quantum Science and Technology (IQST), University of Stuttgart, Stuttgart, Germany

Novel designs for computational devices may offer improvements in energy efficiency, especially if they include a combination of storing and processing parts in one unit. In such designs, the building blocks should enable slow and fast dynamics, and allow for precise control over the system. Atomic-scale frustrated spin systems combined with atom manipulation capabilities of a scanning tunneling microscope (STM) are a promising prospect in this field. Magnetic atoms placed on a thin hexagonal insulator, such as zinc oxide (ZnO), can potentially be used to realize such systems. As the magnetic atoms interact with their substrate, it is important to understand the substrate's influence on the atoms' characteristics. We use multiplet calculations to study the interactions of individual magnetic atoms with the ZnO substrate. With these calculations, we can make predictions about the magnetic characteristics of different atoms placed on top of this hexagonal substrate, in particular their orbital and spin moments. We are particularly interested in finding atomic-scale systems that, combined with atom manipulation capabilities of an STM, allow for further advancements in the field of atomic-scale frustrated spin systems.

O 45.17 Tue 18:00 P2

Exploiting YSR States for Driving Spin Transitions in a Magnetic Field — •Maneesha Ismail[1], Juan Carlos Cuevas[2], and Christian R. Ast[1] — [1]Max-Planck-Institute for Solid State Research, Stuttgart, Germany — [2]Universidad Autónoma de Madrid, Madrid, Spain

In recent years, advances in the field of atomic scale electron spin resonance (ESR-STM) have made it possible to manipulate single spins and observe their interaction [1,2]. In this work, we exploit the tip confinement of superconductivity in a magnetic field to explore a new candidate system, namely Yu-Shiba-Rusinov states. We present how the Zeeman-split states behave under microwave irradiation. Our theory, which is based on a Green's function approach, corroborates the idea that the presence of the YSR state can be used to detect a change in the spin population. A new sample system could open the path to observing new phenomena in the field of spintronics and quantum computing.

1. K. Yang et al., Science 366 (6464), 509-512
2. L. Sellies et al., Nature 624, 64, 2023.

O 45.18 Tue 18:00 P2

Tuning the interaction between spin triplet states at the single-molecule level — Lorenz Meyer, •Maximilian Kögler, Robert Henninger, Nicolas Néel, and Jörg Kröger — Technische Universität Ilmenau, Ilmenau, Germany

In a scanning tunneling microscope junction, two molecular spins - one nickelocene (Nc) decorating the tip and another adsorbed on Pb(111) - are approached toward each other. Simultaneously, inelastic electron tunneling spectra reveal the evolution of spin excitations with increasing magnetic exchange interaction. An avoided energy level crossing occurs at the verge of the Nc-Nc contact. Modeling of the spectra hints at the exchange coupling of tunneling electrons to the Nc spins as the origin. Funding by the DFG through KR 2912/18-1 and KR 2912/21-1 and the BMBF through the ForLab initiative is acknowledged.