O 43: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies IV

Time: Wednesday 10:30–13:00 Location: MA 004

O 43.1 Wed 10:30 MA 004

A scanning quantum sensor for atomic-scale electric and magnetic fields — •Taner Esat[1,2], Dmitriy Borodin[3,4], Jeongmin Oh[3,4], 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 03760, South Korea — [4]Department of Physics, Ewha Womans University; Seoul 03760, 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 report the fabrication of a quantum sensor delivering atomic resolution and single-spin sensitivity, assembled atom-by-atom at the apex of an ultra-high vacuum low temperature scanning tunnelling microscope (STM) tip. Our quantum sensor consists of a magnetic tip with an aromatic molecule attached by atomic-scale manipulation, forming a quantum mechanical two-level system in an external magnetic field. It is addressable by STM-based electron spin resonance and allows readout by tunneling magnetoresistance. We demonstrate the functionality of the quantum sensor by measuring the magnetic and electric dipole fields emanating from a single Fe atom and an Ag2 dimer with sub-microvolt energy and simultaneous atomic-scale spatial resolution.

O 43.2 Wed 10:45 MA 004

Controlling spin excitations of a single molecular magnet — •Maximilian Kögler, Nicolas Néel, and Jörg Kröger — Institut für Physik, TU Ilmenau, Ilmenau, D-98693, Germany

A single molecular magnet (SMM) with a robust spin S=1 was attached to the tip of a scanning tunneling microscope. The spin excitation spectrum was probed by inelastic electron tunneling spectroscopy as a function of the vertical distance to the metal surface, an adsorbed atom and an artificially crafted metal cluster. Above the clean metal surface the SMM spin excitation reduces its energy upon decreasing the distance. Possibly, the magnetic anisotropy energy is affected by relaxations of the junction geometry. In contrast, the excitation spectrum remains invariant atop the adsorbed atoms and vanishes abruptly upon collapsing the tunneling barrier. Funding by the Deutsche Forschungsgemeinschaft through KR 2912/21-1 is acknowledged.

O 43.3 Wed 11:00 MA 004

Spin engineering in artificial atom-molecule hybridsS — •Wantong Huang[1], Máté Stark[1], Paul Greule[1], Daria Sostina[2], Constantin Weeber[1], Jose Gálvez[3], Christoph Sürgers[1], Christoph Wolf[3], Wolfgang Wernsdorfer[1], and Philip Willke[1] — [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

Protecting individual qubits from decoherence and relaxation is one of the crucial challenges for quantum information processing. Both, individual atoms as well as magnetic molecules 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. The resulting magnetic system consisting of an FePc molecule coupled to an Fe atom, forms a mixed spin(1/2,1) Heisenberg antiferromagnet. We show that the strong exchange coupling leads to a ground-state doublet. Thus, this hybrid behaves effectively like a spin-1/2 system which can be driven into electron spin resonance by means of scanning tunneling microscopy. Strikingly, the relaxation time of the hybrid is significantly enhanced compared to pristine FePc reaching 1 μs. Moreover, we show by building dimers of hybrids that their spins can be coupled providing a path to realizing larger structures.

O 43.4 Wed 11:15 MA 004

Paramagnetic resonance of individual organic molecules driven by quantum spin torque — •Stepan Kovarik[1], Richard Schlitz[1,2], Aishwarya Vishwakarma[1], Dominic Ruckert[1], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]Department of Materials, ETH Zurich, Switzerland — [2]Department of Physics, University of Konstanz, Germany

The transfer of angular momentum between the current of spin-polarised electrons and spins of a magnetic material, the so-called spin torque, is used to control the magnetisation of modern magnetic memory devices [1]. Spin torque has also been used to excite magnetic resonance in ferromagnetic devices. [2]. Here, we use spin torque to drive the paramagnetic resonance of a single spin in a pentacene molecule adsorbed on ultrathin MgO with a time-dependent spin current injected from the tip of a scanning tunneling microscope. This novel approach to controlling single spins complements the well-established magnetic resonance driven by a time-dependent electromagnetic field. The spin-torque-driven resonance facilitates the observation of paramagnetic resonance even in regimes where the thermal polarisation of the measured spin is low. The observed signatures provide the first experimental insights into the spin-torque-driven resonance at the quantum level, previously studied only theoretically [3]. [1]D. C. Ralph, M. D. Stiles, JMMM. 320, 1190-1216 (2008). [2]T. Chen et al., Proc. IEEE. 104, 1919-1945 (2016). [3]A. M. Shakirov et al., PRB. 99, 054434 (2019).

O 43.5 Wed 11:30 MA 004

Magnetic resonance imaging of single organic radicals on a surface — •Christoph Wolf[1,2], Gregory Czap[3], Jose Reina-Gálvez[1,2], and Christopher P. Lutz[3] — [1]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, 03760 Republic of Korea — [2]Ewha Womans University, Seoul, 03760 Republic of Korea — [3]IBM Research Division, Almaden Research Center, San Jose, CA, 95120 USA

The combination of electron spin resonance and scanning tunneling microscopy resulted in a versatile surface probe with sub-nm spatial and μeV energy resolution. In this work, we will show that this technique can be applied to characterize singly occupied molecular orbitals of organic radicals. We show the general applicability of this technique by applying it across a series of small fused hydrocarbons adsorbed on thin layers of magnesium oxide used to decouple the spins from the underlying silver substrate. We utilize ab initio calculation and transport simulations using non-equilibriums Green*s functions to explain spatial and spectral variation of the magnetic resonance signal. Our work is the first demonstration of magnetic resonance imaging of a single molecular orbital with sub-molecular resolution of an organic radical of a surface.

O 43.6 Wed 11:45 MA 004

Magnetic resonance imaging of an electron spin residing in an extended molecular orbital — •Richard Schlitz[1,2], Stepan Kovarik[1], Aishwarya Vishwakarma[1], Dominic Ruckert[1], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]Department of Materials, ETH Zürich, 8093 Zürich, Switzerland — [2]Department of Physics, University of Konstanz, 78457 Konstanz, Germany

The combination of a scanning tunneling microscopy (STM) and electron paramagnetic resonance (EPR) allows addressing individual spins on surfaces with sub-μeV energy resolution and pm spatial resolution. Recent works demonstrated that EPR-STM can be used for magnetic resonance imaging (MRI), which reveals the interaction between the magnetic STM tip and the investigated spin species [1]. However, until now imaging was only performed for spins that were localized in an atomic orbital. In this talk, we will show the MRI of pentacene on two monolayers of MgO on Ag(001) by EPR-STM. Pentacene on MgO is singly charge with the electron residing in a delocalized molecular orbital. We discuss how the resonant slice is modified by the extended nature of the electron orbital and critically highlight differences to the atomic systems [2].

[1] Willke et al., Nature Physics 15, 1005-1010 (2019).
[2] Kovarik, Schlitz et al., submitted.

O 43.7 Wed 12:00 MA 004

Single-molecule electron-spin resonance by means of atomic force microscopy — Lisanne Sellies, •Sonja Bleher, and Jascha Repp — University of Regensburg, Regensburg, Germany

Recently, we combined electron spin resonance (ESR) and atomic force microscopy (AFM). To this end we drove electron spin transitions between the non-equilibrium triplet states of single molecules, which differ in their lifetimes. Driving these transitions changes the overall triplet lifetime [1], which can be read out with AFM using an electronic pump-probe scheme [2]. The resulting ESR-AFM spectra exhibit a sub-nanoelectronvolt spectral resolution and allow us to distinguish molecules that only differ in their isotopic configuration, as we demonstrated for pentacene [3].

This high spectral resolution allows us to detect minor influences of the local environment. For example, changing the voltage between the tip and the sample, we detected a considerable Stark shift. This Stark shift together with the cantilever oscillation likely contributes substantially to the small but finite linewidth. In this contribution, recent results obtained by ESR-AFM will be presented.

References:
[1] Köhler (1999). Physics Reports, 310, 261-339.
[2] Peng et al. (2021). Science, 373, 452-456.
[3] Sellies et al. (2022). arXiv, arXiv:2212.12244.

O 43.8 Wed 12:15 MA 004

Three-spin model for Dy atom adsorbed on graphene/Ir(111) — •Jindrich Kolorenc — Institute of Physics (FZU), Czech Academy of Sciences, Prague, Czech Republic

The inelastic electron tunneling spectroscopy (IETS) revealed that rare-earth atoms adsorbed on graphene/Ir(111) carry magnetic moments not only in their 4f shell but also in their valence shell(s) [1]. The measured IETS spectra can be accurately reproduced by assuming that there is one unpaired electron in the 6s orbital (or, more accurately, in some 6s–5d hybrid orbital) [1,2]. Recently, it was argued that to understand the spin dynamics in these adatoms, two separate spin-1/2 moments are needed to be present in the valence shells, probably one in the 6s orbital and one in the 5d orbitals [3]. Using theoretical modeling, we analyze what implications this additional spin has for the IETS spectra. We show that there should be one more step in the $\mathrm{d}I/\mathrm{d}V$ curve in addition to those reported in [1]. Combining first-principles and empirical parameters, we estimate the energy of this additional excitation.

[1] M. Pivetta *et al.*, Phys. Rev. X **10** (2020) 031054
[2] D. Kyvala, J. Kolorenc, `https://www.dpg-verhandlungen.de/year/2023/conference/skm/part/o/session/33/contribution/6`
[3] A. Curcella *et al.*, Phys. Rev. Lett. **130** (2023) 106702

O 43.9 Wed 12:30 MA 004

Electrically Driven Spin Resonance of 4*f* Electrons in a Single Atom on a Surface — •Stefano Reale[1,2,3], Jiyoon Hwang[1,4], Jeongmin Oh[1,4], Harald Brune[5], Andreas J. Heinrich[1,4], Fabio Donati[1,4], and Yujeong Bae[1,4] — [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 Energy, Politecnico di Milano, Milano 20133, Italy — [4]Department of Physics, Ewha Womans University, Seoul 03760, Republic of Korea — [5]Institute of Physics, Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

Lanthanide atoms on surfaces are an exceptional platform for atomic-scale magnetic information storage. However, their potential as qubits is yet unexplored due to the limited number of experimental set-ups that can coherently drive the spins of single adatoms. Through x-ray magnetic circular dichroism and multiplet calculations, we identified erbium (Er) atoms on MgO(100)/Ag(100) as promising candidates for quantum coherent operations. This is due to their magnetic ground state and the inherent decoupling from the environment typical of the 4*f* shell. Here we employed scanning tunneling microscope to drive electron spin resonance (ESR) in a single Er atom. We exploited the well characterized titanium (Ti) atom to engineer a suitable Ti-Er dimer through atomic manipulation. This architecture allows us, through magnetic coupling, to access the spin state of Er, driving and detecting ESR transitions on its elusive 4*f* spins.

O 43.10 Wed 12:45 MA 004

A straight forward method to read the nuclear qudit of 4f-single molecular magnets: ^{163}DyPc$_2$ — •Hongyan Chen[1], Simon Gerber[1], Philip Schmid[1], Vera Schmeiser[1], Svetlana Klyatskaya[1], Eufemio Moreno-Pineda[2], Mario Ruben[1,3], and Wulf Wulfhekel[1] — [1]Karlsruhe Institute of Technology , Germany — [2]Universidad de Panamá — [3]Centre Européen de Sciences Quantiques, Strasbourg

The nuclear spin in single molecular magnets (SMMs) has been used for quantum information processing and has so far been detected by transitions of the magnetic moment during sweeping the magnetic field [1]. We here present a faster method to directly read the nuclear spin (I=5/2) of ^{163}DyPc$_2$ using spin-polarized scanning tunneling microscopy (Sp-STM) without the need for a magnetic field. For (I=0) DyPc$_2$ on Au(111), we recently demonstrated that the Dy^{3+} electron spin can be read out by Sp-STM but loses remanence due to the non-Kramers nature [2]. Adding the nuclear spin of I=5/2 restores Kramers protection and shifts the magnetization curve of Dy^{3+} by the hyperfine field. Measuring the spin polarization of the Kondo state at vanishing magnetic field thus directly reveals the quantum state of the nucleus. We observe T_1 times in excess of minutes at 35 mK. First results on nuclear magnetic resonance transitions between the nuclear states inducing by radio frequency voltages injected into the tunneling junction will be presented. [1] R. Vincent et al,. Nature 488, 357 (2012); S.Thiele et al,. Science 344, 1135 (2014). [2] T. Frauhammer, H. Chen et al,. Phys.Rev.Lett. 127, 123201 (2021).