O 47: Focus Session: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy 1

Magnetic single atoms and molecules are intensively studied as the smallest building blocks for potential applications in spintronic devices and quantum information processing. Detecting and controlling single spin states and their spin interactions require both high energy resolutions and atomic scale imaging capability. Scanning tunneling microscopes provide not only the spatial resolutions but also the bottom up approach to build magnetic structures atom by atom. Recently, unprecedented spin sensitivities have been reached by functionalizing the STM tips or by combining STM with electron spin resonance. The goal of this symposium is to highlight the recent developments in the rapidly evolving field of atomic scale sensing and quantum control of spins on surfaces. Researchers that use STM to investigate atomic or molecular spins on surfaces will present their most recent results. Sharing technical advances and addressing current issues will create synergies to foster future progress in this field and a deeper understanding of the underlying physics.

Organizer: Andreas Heinrich (Center for Quantum Nanoscience, Seoul, Republic of Korea)

Time: Wednesday 15:00–18:00 Location: S051

Topical Talk O 47.1 Wed 15:00 S051

Quantum control of multi-spin architectures on a surface — •Yujeong Bae — Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), 03760 Seoul, South Korea — Department of Physics, Ewha Womans University, Seoul 03760, South Korea

The spin-polarized scanning tunneling microscopy (STM) combined with electron spin resonance (ESR) [1] enables us to achieve single spin sensitivity with atomic precision. Employing ESR-STM, a single spin on a surface can be coherently probed and controlled [1,2], where the magnetic tip is positioned directly on the target spin. Here, we demonstrate a new approach to coherently control multi-electron spins in a quantum spin architecture crafted atom-by-atom. We found the remote spins, which are outside the tunnel junction, can be controlled by the local oscillating magnetic fields created by a single-atom magnet placed next to them in oscillating electric fields. The readout of multi-electron spins is achieved by a sensor atom weakly coupled to them. The resonances of the sensor spin are separated in the frequency domain so that we can independently and simultaneously control the sensor and remote spins. Our work shows the enhanced coherent properties of the remote spins as well as fast controlled operations of multi-electrons in an all-electrical fashion. Our development widens the approaches to the multi-spin control in tailored spin structures on a surface.

[1] S. Baumann et al., Science, 350 (2015) 417.
[2] K. Yang et al., Science, 366 (2019) 509.

O 47.2 Wed 15:30 S051

Longitudinal and transverse electron paramagnetic resonance in a scanning tunneling microscope — •Tom S. Seifert[1], Stepan Kovarik[2], Pietro Gambardella[2], and Sebastian Stepanow[2] — [1]FU Berlin — [2]ETH Zürich

Combining scanning tunneling microscopy (STM) with electron-paramagnetic resonance (EPR) allows for sensitive probing magnetic interactions at atomic scales [1]. However, the experimental requirements for driving the EPR transitions are not obvious [2,3]. In-depth understanding of what drives EPR-STM is mandatory to explore novel material systems with high sensitivity. Here, we acquire and model EPR spectra of single Fe and hydrogenated Ti atoms on bilayer MgO on Ag using a radio frequency (RF) antenna close to the STM junction [4]. We investigate in a systematic way the impact of RF excitation strength and tunneling parameters on the EPR signal and find strong evidence for a piezoelectric coupling mechanism [5]. Specifically, transverse magnetic field gradients drive the spin-1/2 hydrogenated Ti, whereas longitudinal magnetic field gradients drive the spin-2 Fe. Finally, we demonstrate how the choice of specific tip-sample distances allows one to minimize the impact of tip magnetic fields on the EPR-STM measurements thereby excluding a major experimental uncertainty when determining single-atom magnetic moments [6].

[1] S. Baumann et al., Science 350 (2015) [2] K. Yang et al., PRL 122 (2019) [3] P. Willke et al., Nano Lett. 19 (2019) [4] T.S. Seifert et al., PRR 2 (2020) [5] T.S. Seifert et al., Sci. Adv. 6 (2020) [6] T.S. Seifert et al., PRR 3 (2021)

O 47.3 Wed 15:45 S051

Experimental Determination of a Single Atom Ground State Orbital through Hyperfine Anisotropy — •Laëtitia Farinacci[1], Lukas M. Veldman[1], Philip Willke[2], and Sander Otte[1] — [1]Delft University of Technology, Delft, The Netherlands — [2]Karlsruhe Institute of Technology, Karlsruhe, Germany

Electron spin resonance has long been a powerful tool for electronic analysis. Its recent combination with scanning tunneling microscopy [1] gives exceptional energy resolution for the investigation of the magnetism of single atoms in addition to a very good characterization of their spatial surroundings.

Here, we provide a full angle-dependent investigation of the anisotropy of the hyperfine splitting of single Ti atoms on MgO/Ag(100). We find that the anisotropy of the hyperfine splitting is related to that of the g factor: spin-orbit coupling leads to a partially unquenched angular momentum which couples to the electron spin and thereby affects its interaction both with an external magnetic field and the nuclear spin of the nucleus. Combining the symmetry properties of the atoms binding site with a simple point charge model, we provide a method to predict the shape of the ground state orbital of the Ti atom. Relying on experimental values only, this analysis paves the way for a new protocol for electronic structure analysis for spin centers on surfaces.

[1] Baumann et al., Science 350 (2015)

O 47.4 Wed 16:00 S051

Anisotropic hyperfine interaction of surface-adsorbed single atoms — •Jinkyung Kim[1,2], Kyungju Noh[1,2], Yi Chen[1,3], Christoph Wolf[1,3], Andreas Heinrich[1,2], and Yujeong Bae[1,2] — [1]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), Seoul 03760, South Korea — [2]Department of Physics, Ewha Womans University, Seoul 03760, South Korea — [3]Ewha Womans University, Seoul 03760, Republic of Korea

Hyperfine interactions between electron and nuclear spins provide sensitive probes to the chemical environment of atoms, molecules, and crystal defects as well as an alternative way to control the nuclear spins. While a variety of experimental techniques have been applied to the detection and control of single nuclear spins in various environments, simultaneous investigations of the single nuclear spin*s resonance and the atomic scale imaging are more demanding. Using the electron spin resonance (ESR) technique in a scanning tunneling microscope (STM), we investigated hyperfine interaction of hydrogenated titanium (Ti) on MgO/Ag(100) and its local environment at the atomic scale. By means of atom manipulation in vector magnetic fields, we identified the hyperfine interaction of Ti along three principal axes, which shows a large anisotropy of hyperfine interaction. As a sensitive probe of chemical environment, the observed hyperfine interaction reflects the anisotropic orbital configuration of the electronic ground state, which is further supported by the density functional theory calculations.

O 47.5 Wed 16:15 S051

Electron Paramagnetic Resonance of individual Alkali Metal Atoms and Dimers on Ultrathin MgO — •Stepan Kovarik[1], Roberto Robles[2], Richard Schlitz[1], Tom Sebastian Seifert[1,3], Nicolas Lorente[2,4], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]Department of Materials, ETH Zurich, Switzerland — [2]Centro de Física de Materiales, San Sebastian, Spain — [3]Department

of Physics, FU Berlin, Germany — [4]Donostia International Physics Center, San Sebastian, Spain

Electron paramagnetic resonance (EPR) provides unique insight into the chemical structure and magnetic properties of dopants in oxide and semiconducting materials that are of interest for applications in electronics, catalysis, and quantum sensing. We demonstrate that EPR in combination with scanning tunneling microscopy (STM) [1, 2] allows for probing the spin and charge state of alkali metal atoms on an ultrathin magnesium oxide layer on a Ag(100) substrate. We identify a magnetic moment of 1 μ_B for Li_2, LiNa, and Na_2 dimers corresponding to spin radicals with a charge state of +1e [3]. Individual alkali atoms have the same charge state and no magnetic moment. The ionization of the adsorbates is attributed to the charge transfer of one electron through the oxide to the metal substrate. Our work highlights the potential of EPR-STM to provide insight into dopant atoms, which are relevant for the control of the electrical properties of surfaces and represent a suitable platform for studying quantum nanomagnets.

[1] S. Baumann et al., Science 350 (2015) [2] T. S. Seifert et al., PRR 2 (2020) [3] S. Kovarik et al., Nano Lett. 22 (2022)

Topical Talk O 47.6 Wed 16:30 S051

Free coherent evolution of a coupled atomic spin system initialized by electron scattering — •Sander Otte[1], Lukas Veldman[1], Laetitia Farinacci[1], Rasa Rejali[1], Rik Broekhoven[1], Jeremie Gobeil[1], David Coffey[1], and Markus Ternes[2,3] — [1]Delft University of Technology, Delft, The Netherlands — [2]RWTH Aachen University, Aachen, Germany — [3]Forschungszentrum Jülich, Jülich, Germany

Full insight into the dynamics of a coupled quantum system depends on the ability to follow the effect of a local excitation in real-time. Here, we trace the free coherent evolution of a pair of coupled atomic spins by means of scanning tunneling microscopy. Rather than using microwave pulses, we use a direct-current pump-probe scheme to detect the local magnetization after a current-induced excitation performed on one of the spins. By making use of magnetic interaction with the probe tip, we are able to tune the relative precession of the spins. We show that only if their Larmor frequencies match, the two spins can entangle, causing angular momentum to be swapped back and forth. These results provide insight into the locality of electron spin scattering and set the stage for controlled migration of a quantum state through an extended spin lattice.

O 47.7 Wed 17:00 S051

Controlled migration of a coherent spin excitation through atomically assembled nanomagnets — •Lukas Veldman[1], Laëtitia Farinacci[1], Rasa Rejali[1], Rik Broekhoven[1], Jeremie Gobeil[1], David Coffey[1], Markus Ternes[2,3], and Sander Otte[1] — [1]TU Delft, Delft, The Netherlands — [2]RWTH Aachen, Aachen, Germany — [3]Forschungszentrum Jülich, Jülich, Germany

Tracing single coherent spin excitations in low dimensional nanomagnets has been a long standing goal in experimental solid state physics. Scanning tunneling microscopy offers a promising platform to chase this goal due to its capability to build nanomagnets atom-by-atom and address each atomic spin individually. Here, we show the possibility of inducing a single spin-flip excitation using electron scattering and the measurement of the resulting flip-flop interaction between two atomic spins [1]. Next, we use the same principle to observe coherent spin dynamics in multiple atomically assembled nanomagnets. By tuning the interaction between tip and nanomagnet, we are able to address different magnetic resonances in each atomic structure. In engineered branched structures, the spin excitation can be sent to different directions using the tip interaction. These techniques can serve as a platform for dynamical quantum simulation and can form a foundation for atomically assembled spintronic applications.

[1] Veldman, L. M., Farinacci, L., Rejali, R., ... & Otte, A. F. (2021) Science, 372

O 47.8 Wed 17:15 S051

Electric field control of spin transitions in a single molecule using ESR-STM — •Maneesha Ismail, Piotr Kot, and Christian R. Ast — Max-Planck-Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany

Since the demonstration of the control of spins on the atomic scale, the technique of ESR-STM has been used extensively to explore the field of spintronics. Here, we present an extension to the ESR-STM parameter space, which uses the bias voltage to tune the energy of the Zeeman transition. We emonstrate electronic control of spin resonance transitions in a single TiH molecule. We were able to observe a strong dependency of the g-factor along with a tip-field shift as function of the electric field. Finally, we manipulate a TiH dimer by continuously changing the bias voltage such that the dimer moves through an avoided crossing of the energy levels. This could be an important step towards pump-probe control of spin states through the bias voltage and opens new possibilities for coherent manipulation.

O 47.9 Wed 17:30 S051

Isotope detection inside single molecules in scanning-probe based electron spin resonance — •Lisanne Sellies, Raffael Spachtholz, Philipp Scheuerer, and Jascha Repp — University of Regensburg, Regensburg, Germany

Electron spin resonance (ESR), a versatile technique to study materials with unpaired electrons, was recently combined with scanning tunneling microscopy (STM), bringing atomic-scale spatial resolution to ESR [1]. ESR-STM even allowed detecting strong hyperfine interactions between the single electron spin under study with the atom's nuclear spin [2], due to its largely improved energy resolution compared to conventional STM, reaching the nano-electron-volt regime. This energy resolution could even be improved further if the tunneling current as a read-out was avoided, since this current is the dominating decoherence source for the probed electron spin.

We propose, therefore, a new ESR scanning probe method based on atomic force microscopy (AFM). Since our technique does not rely on the tunneling current to read-out the ESR signal, we increase the coherence times of the electron spins and, consequentially, the energy resolution. Therefore, we can resolve the hyperfine interaction inside organic molecules and distinguish molecules only differing in the isotopic composition. Thus, our technique allows the chemical fingerprinting of molecules and their surroundings.

References: [1] S. Baumann et al., Science 350, 417 (2015) [2] P. Willke et al., Science 362, 336 (2018)

O 47.10 Wed 17:45 S051

A home-built scanning tunneling microscope combined with electron spin resonance — •Andreas Heinrich — Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Korea

Scanning tunneling microscopy is a powerful tool to characterize the electronic and magnetic properties of atomic scale structures on a surface. Recently, improved spectral energy resolution has been achieved by functionalizing the STM tip with a well-characterized molecule at the apex [1,2] or by combining electron spin resonance with STM (ESR-STM) [3]. Here, we present the design and operation of an optimized, home-built ESR-STM with a specially designed mechanical damper, a Joule-Thomson refrigerator, and 2-axes vector magnets. This system provides outstanding performance of STM for nanoscale measurements. We further describe a new design of a microwave antenna to increase the transmission of RF voltages to the junction, which performs better than the direct connection to the tip. Applying RF power through the antenna terminated at 50 Ohm results in the reduction of standing waves and increases the available frequency range (5-40 GHz), which allows us to measure ESR of surface spins at elevated temperatures up to 10 K. References: [1] M. Ormaza et al., Nano let. 17, 1877-1882 (2017) [2] G. Czap et al., Science, 364, 670-673 (2019) [3] S. Baumann et al., Science, 350, 417-420 (2015)