O 21: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies III

Time: Tuesday 10:30–13:15 Location: MA 004

Topical Talk O 21.1 Tue 10:30 MA 004

Theory of Electron Spin Resonance in Scanning Tunneling Microscopy — •Juan Carlos Cuevas[1], Christian R. Ast[2], Piotr Kot[2], Maneesha Ismail[2], Sebastián de-la-Pena[1], and Antonio I. Fernandez-Dominguez[1] — [1]Departamento de Fisica Teorica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autonoma de Madrid, 28049 Madrid, Spain — [2]Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

Electron spin resonance (ESR) spectroscopy in scanning tunneling microscopy (STM) has enabled probing the electronic structure of single magnetic atoms and molecules on surfaces with unprecedented energy resolution. Despite this remarkable success, the field could still greatly benefit from a more quantitative understanding of the ESR-STM physical mechanisms. Here, we present a theory of ESR-STM which quantitatively models not only the ESR signal itself, but also the full background tunneling current, from which the ESR signal is derived. We show that this theory is able to quantitatively reproduce the experimental results for a spin 1/2 system (TiH molecules on MgO) across many orders of magnitude in tunneling current, providing access to the relaxation and decoherence rates that govern the spin dynamics due to intrinsic mechanisms and to the bias voltage. More importantly, and with the help of additional electromagnetic simulations, our work establishes that the transitions in our ESR-STM experiments can be driven by the ac magnetic field at the junction.

O 21.2 Tue 11:00 MA 004

Coherent spin dynamics between electron and nucleus within a single atom — •Lukas M. Veldman[1,2], Evert W. Stolte[1], Mark P. Canavan[1], Rik Broekhoven[1], Philip Willke[3], Laëtitia Farinacci[1], and Sander Otte[1] — [1]Delft University of Technology, Delft, The Netherlands — [2]University of Stuttgart, Stuttgart, Germany — [3]Karlsruhe Institute of Technology, Karlsruhe, Germany

The study of single atom electron spins has led to both fundamental insight into the building blocks of matter as well as shown the tantalizing potential to construct functional magnetic devices atom-by-atom. The nuclear spin forms the next frontier in this effort, offering insight into intra-atomic magnetic interactions while, at the same time, promising prolonged coherence times for coherent quantum operations compared to their electronic counterparts. Here, we present coherent dynamical access to a single nuclear spin by combining electron spin resonance measurements with direct-current pump-probe spectroscopy inside a scanning tunneling microscope. We initialize both spins by means of the spin polarized tunneling electrons and control the resulting dynamics by tuning the amount of entanglement between nuclear and electron spin with the magnetic probe tip. This local spin control provides a pathway towards utilizing single nuclei for dynamic quantum simulation in extended atomic arrays.

O 21.3 Tue 11:15 MA 004

Single-shot measurement of the nuclear spin state of a single atom using ESR-STM — •Jinwon Lee[1], Evert W. Stolte[1], Hester Vennema[1], Rik Broekhoven[1], Esther Teng[1], Philip Willke[2], and Sander Otte[1] — [1]Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands — [2]Physikalisches Institut, Karlsruhe Institute of Technology, 67131 Karlsruhe, Germany

Individual nuclear spins have arisen as promising candidates for the building blocks for quantum memory because they have longer lifetime and coherence time compared to electronic spin states. Most studies on individual nuclear spins have focused on the nuclear spins embedded in solids and single-molecule magnets, which have limited controllability due to their environment. Scanning tunneling microscopy with electron spin resonance (ESR-STM), which allows for precise placement of individual atoms on a surface, recently observed the nuclear spin state through the hyperfine interaction. However, time-resolved measurements for its relevant timescales have not been reported. In this work, we achieve single-shot measurements of the nuclear spin state of ^{49}Ti atom with S=1/2 and I=7/2, adsorbed on MgO/Ag using ESR-STM. We apply continuous-wave RF electric field, which can drive ESR only when the atom has a certain nuclear spin state and observe whether ESR is driven or not by measuring tunneling conductance. This new approach enables time-resolved measurements of the nuclear spin state, and we measure its dwell time to be on the order of 100 ms, 6 orders of magnitude longer than the electronic spin in the same atom.

O 21.4 Tue 11:30 MA 004

Unveiling second harmonic resonances in ESR/STM Setups using a time dependent Anderson impurity model. — •Jose Reina Galvez[1], Roberto Robles[2], Nicolas Lorente[2], and Christoph Wolf[1] — [1]Center for Quantum nanoscience, Seoul, South Korea — [2]Centro de Fisica de Materiales, San Sebastian, Spain

The achievement of reproducible single-atom and single-molecule electron spin resonance (ESR) in a scanning tunneling microscope (STM) junction opened new possibilities in the analysis of surface science at the atomic scale [1,2]. The predominant feature of a spin driven on resonance is a resonance at the first harmonic of the Larmor frequency in the current spectrum. Higher harmonic excitations of the spin precession can emerge as long as the driving is sufficiently strong due to nonlinearity in the transport. This leads to a 2nd harmonic resonance at half the Larmor frequency.

In this talk, I will discuss the theoretical aspects of the phenomenon above. Our framework utilizes an open quantum systems formalism for a modified Anderson impurity Hamiltonian where the quantum impurity is connected by time dependent hopping terms to two fermionic baths, a polarized STM tip and a metal substrate [3,4,5]. This barrier modulation in the tunneling allows for efficient driving of the spin-1/2 and the emergence of a 2nd harmonic at driving amplitudes VRF in good agreement with the experiment.

References: [1] S. Baumann et al, Science (2015). [2] S. Kovarik et al, Nano Lett. (2022) [3] J. Reina-Gálvez et al, PRB (2021) [4] J. Reina-Gálvez et al, PRB (2023) [5] https://github.com/qphensurf/

O 21.5 Tue 11:45 MA 004

DC-bias gating of single spin electron paramagnetic resonance — Hong T. Bui[1,2], We-hyo Seo[1,2], Valeria Sheina[1,2], and •Soo-hyon Phark[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Korea — [2]Ewha Womans University, Seoul, Korea

Ti and Fe atoms in the tunnel junction of a scanning tunneling microscope combined with electron paramagnetic resonance (EPR) are prototypical quantum systems to study single spin dynamics. However, influence of DC electric bias (Vdc), followed by the spin-polarized current passing through the spins, on the EPR signal have not been studied in depth yet. Here, we introduce DC-bias gating of EPR resonance observed from single Ti and Fe adsorbates on ultrathin MgO layers. We find that a negative Vdc, inducing reversed magneto-tunneling transport, considerably suppresses EPR of Ti and accompanies a reversal of peak asymmetry. This is even more pronounced in the EPR of Fe, where the sign of the peak is inversed with a considerable intensity. Simulations on spin-dependent electron transport through a RF-driven single spin reproduce the experiments on both Ti and Fe in good agreement, revealing that DC-bias-driven spin pumping via the inelastic channels plays a key role in the RF-driven spin dynamics of the EPR-relevant two levels. In addition, a long-lived inversed population in a Fe spin can promote EPR resonance with a reversed sign as observed in the measured spectra.

O 21.6 Tue 12:00 MA 004

Pulse scheme to create and detect entanglement in surface spin systems using a tunneling microscope — •Rik Broekhoven[1], Curie Lee[2,3], Soo-hyon Phark[2,4], Sander Otte[1], and Christoph Wolf[2,4] — [1]Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, the Netherlands — [2]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul 03760, Korea — [3]Department of Physics, Ewha Womans University, Seoul 03760, Korea — [4]Ewha Womans University, Seoul 03760, Korea

Being able to certify quantum entanglement is an essential element of quantum-coherent applications which has not yet been shown in spins on a surface. Here, we present a way to unambiguously identify entanglement by exploiting that entangled states undergo a free evolution with a characteristic time constant different from any other evolution in the system. Our approach is compatible with multi-spin resonance

control in a scanning tunneling microscope (STM). We suggest a pulse scheme that first creates an entangled spin pair outside of the STM junction and subsequently projects the accumulated phase during free evolution on the population of one of the spins, which in turn can be read out through a weakly coupled sensor spin. The resulting measurement signal is an oscillation in the STM current at a time scale proportional to the spin pair exchange coupling if and only if the pair was entangled. We show that this scheme can be implemented using realistic simulation parameters and study the expected measurement contrast as a function of temperature and decoherence times.

O 21.7 Tue 12:15 MA 004

Syncing of stochastically switching atomic orbital memory — •Kira Junghans[1], Hermann Osterhage[1], Werner M. J. van Weerdenburg[1], Ruben Christianen[1], Eduardo J. Domínguez Vázquez[2], Hilbert J. Kappen[2], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University Nijmegen, the Netherlands — [2]Donders Institute for Neuroscience, Radboud University Nijmegen, the Netherlands

Stochastically fluctuating multi-well systems are a promising route toward designing neuromorphic hardware. One challenge is finding tunable material platforms that exhibit such multi-well behavior.
The recently discovered atomic Boltzmann machine provides such a platform, where orbital memory states represent binary stochastic units [1,2]. Here, we investigate the response of orbital memory states of Fe and Co atoms on black phosphorus [2,3] to a sinusoidal input voltage using scanning tunneling microscopy. For both species, we observe synchronization of the state occupation to the sinusoidal signal. The state favorability of Fe atoms also shows a frequency-dependent response to the drive, which can be tuned by the input parameters. In contrast to Fe, there is no significant frequency-dependence in the state favorability for Co atoms. The response of the state favorability can be traced to the difference in the voltage-dependent switching rates of the two species based on the Poisson process model.
[1] B. Kiraly et al., Nat. Nanotechnol. 16, 414 (2021).
[2] B. Kiraly et al., Nat. Commun. 9, 3904 (2018).
[3] B. Kiraly et al., Phys. Rev. Research 4, 33047 (2022).

O 21.8 Tue 12:30 MA 004

Manipulation of Orbital States on Titanium Atoms on MgO/Ag(100) — •Susanne Baumann[1], Daria Sostina[2], Lukas Arnhold[1], Fernando Delgado[3], Philip Willke[2], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Karlsruhe Institute of Technology, Physikalisches Institut, Karlsruhe, Germany — [3]Instituto de estudios avanzados IUDEA, Departamento de Física, Universidad de La Laguna, Spainh

The orbital and spin properties of quantum systems at the nanoscale are heavily influenced by their interaction with the surrounding environment. For atoms adsorbed on a surface, this interaction commonly leads to a partial suppression of the orbital moment, a phenomenon known as orbital quenching. In this study, we investigate and modify the orbital quenching of titanium atoms (Ti) on two monolayers of MgO/Ag(100), the experimental "fruit fly" system for electron spin resonance measurements in conjunction with scanning tunneling microscope (STM). Our study demonstrates how we can finely adjust the spin and orbital excitations by the proximity of the STM tip, ranging from a nearly unquenched orbital moment to a highly quenched state. In addition, we use multiplet and transport calculations to gain a deeper understanding of the intricate interplay between atom adsorption on a surface and the quenching of their orbital moments.

O 21.9 Tue 12:45 MA 004

From single electron ratio rules to atoms on surfaces – A relativistic investigation of hyperfine splittings — •Katharina Lorena Franzke, Wolf Gero Schmidt, and Uwe Gerstmann — Paderborn University, Warburger Str. 100, 33098 Paderborn

The hyperfine splitting of a given system is caused by the interaction of the electronic spin with the magnetic moments of the nuclei and leads to comparably small energy shifts in the absorption spectra. These shifts can be split into the isotropic Fermi contact, the anisotropic dipolar and the orbital part. In many systems, especially in semiconductors, the hybridization and deformation of the atomic orbitals by the crystal field leads to the orbital moment being no longer well defined, but effectively averaged out. This so-called orbital quenching occurs in many, but not all physical systems. We have developed a non-perturbative relativistic method which allows to calculate the contribution of the orbital part for complex structures [1]. We show that the orbital part actually scales with spin-orbit coupling if orbital quenching is hindered by local symmetry, i.e. in case of dimers or atoms at surfaces. This holds true in particular when the unpaired electron is localized in quasi-atomic p-like orbitals. Here, the orbital part is by far not negligible, but becomes dominant by surpassing the dipolar contribution by a factor of five. It is thus required to achieve overall accuracy in predicting the hyperfine interactions.

[1] Franzke K.L. *et al.*, *Relativistic calculation of the orbital hyperfine splitting in complex microscopic structures*, accepted in J. Phys. Com., 2023

O 21.10 Tue 13:00 MA 004

Atomic scale study on electronic configurations of single titanium atoms on MgO — •Hong T. Bui[1,2], We-hyo Seo[1,2], Curie Lee[1,2], Christoph Wolf[1,2], Nicolas Lorente[3,4], and Soo-hyon Phark[1,2] — [1]center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Korea — [2]Ewha Womans University, Seoul, Korea — [3]Centro de Física de Materiales, Donostia-San Sebastián, Spain — [4]Donostia International Physics Center, Donostia-San Sebastián, Spain

A single Ti adsorbate on two-monolayer (ML) MgO on Ag(100) is a prototypical electron spin two-level system (S = 1/2), which has recently been used to demonstrate quantum gates in on-surface atomic-scale qubit platforms using scanning tunneling microscopy (STM) combined with an electron spin resonance (ESR) [1]. Further advanced multiple-order quantum gates, however, are limited by its short coherence mainly due to the insufficient decoupling from the substrate. In this work, we investigated spin-dependent electronic properties of single Ti adsorbates on a 3-ML MgO using tunneling and ESR spectroscopies. Measurements on Ti on an O-O bridge site show a differential conductance similar to that on the same adsorption site of the 2-ML MgO and reveal S = 1/2, a promising candidate of on-surface spin qubit but with longer coherence. On the other hand, Ti adsorbed on an O-atop, in contrast to that on the 2-ML MgO (S = 1/2), shows inelastic tunneling features and uniaxial out-of-plane magnetic anisotropy, a signature of S > 1/2, which are well supported by DFT and multiplet calculations for the electronic ground state of 3d2 and S = 1.