O 15: Spins on Surfaces at the Atomic Scale II

Time: Monday 15:00–18:00 Location: REC C 213

O 15.1 Mon 15:00 REC C 213

Electron-spin resonance atomic force microscopy — •Lisanne Sellies, Raffael Spachtholz, Philipp Scheuerer, and Jascha Repp — University of Regensburg, Regensburg, Germany

Recently, we combined the high energy resolution of electron spin resonance (ESR) with the spatial resolution offered by atomic force microscopy (AFM). This ESR-AFM technique relies on driving electron spin transitions between the non-equilibrium triplet state levels of a single molecule. Since these triplet states typically have different lifetimes, driving such transitions modifies the overall triplet lifetime [1,2], which can be detected by an electronic pump-probe scheme [3].

ESR-AFM allows to measure ESR signals with a sub-nanoelectronvolt energy resolution, as we demonstrated for pentacene on thick NaCl films. Thereby, molecules only differing in their isotopic configuration can be distinguished. Moreover, due to the minimally invasive nature of the ESR-AFM technique, the electron spins of pentacene can be coherently manipulated over tens of microseconds. After introducing ESR-AFM, recent results obtained by this technique will be presented.

References: [1] Köhler et al. (1993). Nature, 363(6426), 242-244. [2] Wrachtrup et al. (1993). Nature, 363(6426), 244-245. [3] Peng et al. (2021). Science, 373(6553), 452-456.

O 15.2 Mon 15:15 REC C 213

Coherent spin dynamics in a single atom hyperfine system — •Lukas Veldman, Laëtitia Farinacci, Evert Stolte, Mark Canavan, and Sander Otte — TU Delft, Delft, The Netherlands

The combination of electron spin resonance (ESR) with scanning tunneling microscopy (STM) has opened up the possibility to investigate the interaction between electrons and nuclei in individual atoms on a surface [1]. Recently, we showed that for TiH atoms on MgO the hyperfine interaction is highly anisotropic and contains information on the groundstate orbital of the atom [2]. In this work, we go to the low field regime where electron and nuclear states hybridize. We use DC voltage pulses [3] to spin pump both the electron and the nuclear spin [4]. We measure the coherent oscillations of the electron spin driven by the hyperfine interaction. These measurements open the door to controlled spin manipulation of nuclear spins in single atoms.

[1] Willke et al. Science 362 (2018)
[2] Farinacci et al. Nano Letters (2022)
[3] Veldman et al. Science 372 (2021)
[4] Yang et al. Nature Nanotechnology 13 (2018)

O 15.3 Mon 15:30 REC C 213

Quantum coherence of single atomic S = 1/2 spins on ultra-thin insulating layers — Hong Bui[1,2], We-Hyo Seo[1,2], Yu Wang[1,2], Andreas Heinrich[1,2], and •Soo-Hyon Phark[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul 03760, Korea — [2]Department of Physics, Ewha Womans Universtiy, Seoul 03760, Korea

A scanning tunneling microscope (STM), when combined with electron spin resonance (ESR), provides a solid-state platform of atomic scale spin qubit. However, its quantum coherence is considerably limited by couplings with metallic substrate and tunneling electrons. Here we report a study on the quantum coherence of single Ti atomic spins (S = 1/2) on two- (2ML) and three- (3ML) monolayer-thick MgO on Ag(100) using pulsed-ESR equipped in a STM at 0.4 K. We were able to drive the coherent rotations of the spin on the 3ML-MgO with a Rabi rate (Ω) up to 100 MHz, which is an order faster than that on the 2ML-MgO. Coherence time (T2) from Hahn-echo measurement revealed the role of the tunneling electrons, resetting the state of a Ti spin with a probability of 0.15/electron, which is the same for Ti spins on both 2ML- and 3ML-MgO. Of more interesting is the zero current T2 of 2 microsec on 3ML-MgO, about 5 times longer than that on 2ML-MgO, emphasizing the importance of decoupling the spin from the metallic substrate as well as tunneling electrons. Such considerable improvements in Ω and T2 are promising for high fidelity quantum gate operations to a Ti spin qubit, further brightening a way to raise the on-surface spin qubits on a stage of the quantum-coherent applications.

O 15.4 Mon 15:45 REC C 213

Certifying entanglement of spins on surfaces using ESR-STM — •Yelko del Castillo[1,2] and Joaquín Fernández-Rossier[1] — [1]International Iberian Nanotechnology Laboratory (INL), Av. Mestre José Veiga, 4715-330 Braga, Portugal — [2]Centro de Física das Universidades do Minho e do Porto, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal

We propose a protocol to certify the presence of entanglement in artificial on-surface atomic and molecular spin arrays using electron spin resonance carried by scanning tunnel microscopes (ESR-STM). We first generalize the theorem that relates global spin susceptibility as an entanglement witness to the case of anisotropic Zeeman interactions, relevant for surfaces. We then propose a method to measure the spin susceptibilities of surface-spin arrays combining ESR-STM with atomic manipulation. Our calculations show that entanglement can be certified in antiferromagnetically coupled spin chains using state of the art spectral resolution of ESR-STM magnetometry.

O 15.5 Mon 16:00 REC C 213

Ultrafast dynamics of magnetic adatoms on MgO — •Lukas Arnhold[1], Daria Sostina[2], Nicolaj Betz[1], Susanne Baumann[1], Philip Willke[2], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Karlsruhe Institute of Technologies, Physikalisches Institut, Karlsruhe, Germany

The utilization of single magnetic atoms as qubits requires detailed understanding of their energy levels and the associated time-scales, ideally also those of excited states. Ever since coherent driving of a single spin in the junction of a scanning tunneling microscope has been shown [1], the technique of ESR-STM has been established and used for example to perform qubit operations, as well as, to address multiple qubits [2]. Besides the technique of electron spin resonance, quantum stochastic resonance measurements offer new insights into a system's energy transition cycles, or in other words, it's associated time-scales [3]. Applying this measurement scheme onto ESR active adatoms on a MgO substrate, gives access to additional manipulation pathways and allows for further understanding and mastery of coherent spin manipulation.

[1] Baumann, Susanne et al. 'Electron paramagnetic resonance of individual atoms on a surface.' Science 350 (2015): 417 - 420.

[2] Phark, Soo-Hyon et al. 'Double Electron Spin Resonance of Engineered Atomic Structures on a Surface.' arXiv:2108.09880, 2021.

[3] Haenze, Max et al. 'Quantum Stochastic Resonance of individual Fe atoms.' Sci. Adv. Vol 7, Issue 33 (2021).

O 15.6 Mon 16:15 REC C 213

All-electrical driving and probing dressed states of a single spin on surface — •Hong Bui Thi[1,2], Christoph Wolf[1,3], Yu Wang[1,3], Masahiro Haze[1,4], Yi Chen[1,3], Yujeong Bae[1,2], Andreas Heinrich[1,2], and Soo-Hyon Phark[1,3] — [1]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul 03760, Korea — [2]Department of Physics, Ewha Womans University, Seoul 03760, Korea — [3]Ewha Womans University, Seoul 03760, Korea — [4]The Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan

We demonstrate the creation of dressed states of a single electron spin localized on a surface by using a resonant radio-frequency field in a scanning tunneling microscope. Owing to the unique sub-nm probe-sample geometry field strengths as high as 1 GV/m can be achieved at a bias of the radio-frequency field below 100 mV. Magnetic resonance of a single spin in such a cavity is a direct consequence of coupling between the atomic dipole moment and driving electric field, resulting in the dressing of the atomic states. The read-out of these dressed states is facilitated in an all-electrical fashion by a weakly coupled probe spin. Our work highlights the strength of the atomic-scale geometry inherent to the scanning tunneling microscope in terms of the creation and control of dressed states, which are known to provide decoherence-free subspaces critical to the creation of long-lived quantum states.

O 15.7 Mon 16:30 REC C 213

Surface spins as quantum bits - a simulation perspective — Jose Reina Gálvez[1], Nicolas Lorente[2], and •Christoph Wolf[1] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, South Korea — [2]Materials Physics Center, San Sebastian, Spain

Spins localized at or close to a clean surface have emerged as systems of interest in quantum nanoscience. They can be tailored to a desired spin state, manipulated to form structures with designed interactions and as of recent have been used to implement basic quantum logic operations - a control-NOT gate - in a scanning tunneling microscope (STM). In this work we want to explore quantum coherent phenomena of surface spins from a computational perspective. Since surface spins interact rather strongly with the environment, we performed open quantum systems calculations. First, we will show how electron-electron double resonance can be understood by solving the time-dependent Schrodinger equation under continuous and pulsed radio-frequency driving. Using the same prototypical system of two coupled electron spins we also show how the AC Stark effect manifests itself as Autler-Townes doublets and Mollow triplets. To go one step further, we use our recently developed approach to explicitly calculate the current present in the STM experiment using the non-equilibrium Green's functions formalism. These simulations show how the current plays a dual role in these systems by driving quantum transitions but also limiting quantum coherence. Our simulations outline realistic expectations for quantum coherent surface spin systems and their prospective applications.

O 15.8 Mon 16:45 REC C 213

A non equilibrium Green's Function formalism applied to STM/ESR experiments — •Jose Reina-Galvez[1], 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 demonstration of reproducible single-atom and single-molecule electron spin resonance (ESR) in a STM junction opened new possibilities in the analysis of surface science at the atomic scale. Recent experiments have demonstrated quantum-coherent Rabi oscillations and even the implementation of a quantum logic operation: a CNOT gate.

Here, we want to focus on the theory we developed to reproduce some relevant results in the field. Our theoretical framework uses open quantum systems formalism in combination with non equilibrium Green's Function technique. It consists of a quantum impurity connected to a polarized STM tip and an isolated surface, both considered as fermionic baths. The time dependent bias voltage used in the experiments is introduced in the hopping terms. These hoppings are driving the electron transitions from the baths to the quantum impurity which uses a modified Anderson impurity model with localized electronic states, a Coulomb repulsion term and a Zeeman term.

Our model is general, allowing to connect an arbitrary number of localized spins with the transport spin. We will demonstrate how driving the system on resonance modifies the population and the calculated true current through the many-body impurity which can be directly compared to the experiment.

O 15.9 Mon 17:00 REC C 213

Microscopic theory of spin-relaxation of a single Fe adatom coupled to substrate vibrations — •Haritz Garai-Marin[1,2], Manuel dos Santos Dias[3,4,5], Samir Lounis[3,4], Julen Ibañez-Azpiroz[6,7], and Asier Eiguren[1,2,8] — [1]Physics Department, University of the Basque Country UPV/EHU, Bilbao, Spain — [2]Donostia International Physics Center, Donostia, Spain — [3]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich & JARA, Jülich, Germany — [4]Faculty of Physics, University of Duisburg-Essen & CENIDE, Duisburg, Germany — [5]Scientific Computing Department, STFC Daresbury Laboratory, Warrington, United Kingdom — [6]Centro de Física de Materiales CSIC-UPV/EHU, Donostia, Spain — [7]IKERBASQUE Basque Foundation for Science, Bilbao, Spain — [8]EHU Quantum Center, University of the Basque Country UPV/EHU

Understanding the spin-relaxation mechanism of single adatoms is an essential step towards creating atomic magnetic memory bits or quantum bits. Here we present a theory to study the role of electron-phonon coupling by combining *ab-initio* electronic and vibrational properties with the multiplet many-body nature of atomic states. Our calculations reproduce the millisecond spin lifetime measured on Fe adatoms on MgO/Ag(100), which demonstrates that the essential features of the spin-phonon coupling are successfully captured. We show a clear fingerprint for experimentally detecting a localized spin-phonon excitation and we show how the atomic interaction with the environment should be tuned in order to enhance the magnetic stability.

O 15.10 Mon 17:15 REC C 213

Electron spin resonance of individual magnetic molecules on surfaces — •Wantong Huang[1], Máté Stark[1], Paul Greule[1], Daria Sostina[2], David Collomb[1], Christoph Sürgers[1], 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

Single molecular magnets which consist of one central metal ion surrounded by ligands are promising building blocks for nanoscale spintronics and future quantum devices. The recent development of scanning tunnelling microscopy (STM) combined with electron spin resonance (ESR) constitutes here a powerful technique to image and coherently control individual molecular spins on surfaces [Zhang et. al, Nat. Chem. 14, 59 (2022); Willke, ACS Nano, 15, 11, 17959 (2021)]. Here, we explore the magnetic properties and spin dynamics of iron phthalocyanine (FePc) molecules adsorbed on magnesium oxide (MgO) grown on Ag(100). We investigate the magnetic properties of different FePc by scanning tunneling spectroscopy and ESR-STM. Such molecular magnetic structures provide a versatile platform to study complex magnetic interactions and atomic-scale spin dynamics.

O 15.11 Mon 17:30 REC C 213

Orbital and spin excitations of hydrogenated titanium modulated by electric field — •Daria Sostina[1], Lukas Arnhold[2], Nicolaj Betz[2], Susanne Baumann[2], Philip Willke[3], Wolfgang Wernsdorfer[3], and Sebastian Loth[2] — [1]Institute of Quantum Materials and Technologies (IQMT), Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [3]Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany

The goal for coherent control of individual spins on surfaces has prompted the development of new local probe techniques such as a combination of electron spin resonance (ESR) with scanning tunneling spectroscopy (STM). Yet, the excitation mechanism of ESR in STM is not conclusively determined. [Baumann et.al, Science 350, 6259, 2015, Ferron et. al, Phys. Rev. Research 1, 033185 (2019)]. Here we study a higher order orbital excitation of hydrogenated titanium complexes positioned on the oxygen site of the MgO surface. We observe a strong dependence of its excitation energy on the electric field generated by the STM tip. At the same time an ESR transition on that TiH exhibits a shift with the DC bias voltage applied to the junction which was attributed to a change in g-factor of the complex [Kot et. al, arXiv 2209.10969, 2022]. The ability to tune orbital and spin excitations by means of electric fields gives further insight on new pathways for electrical control of single spins centers on surfaces.

O 15.12 Mon 17:45 REC C 213

Lanthanide atoms on MgO(100)/Ag(100) as Candidate for Single-Atom-Qubits — Stefano Reale[1,2,3], Aparajita Singha[1,2,4], Safa L. Ahmed[1,2], Denis Krylov[1,2], Luciano Colazzo[1,2], Christoph Wolf[1,2], Carlo S. Casari[3], Alessandro Barla[5], Edgar Fernandez[6], Francois Patthery[6], Marina Pivetta[6], Stefano Rusponi[6], Harald Brune[6], and •Fabio Donati[1,2] — [1]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), Republic of Korea — [2]Department of Physics, Ewha Womans University, Republic of Korea — [3]Department of Energy, Politecnico di Milano, Italy — [4]Max Planck Institute for Solid State Research, Germany — [5]Istituto di Struttura della Materia (ISM), Consiglio Nazionale delle Ricerche (CNR), Italy — [6]Institute of Physics, Ecole Polytechnique Federale de Lausanne, Switzerland

Lanthanide atoms on surfaces are an exceptional platform for atomic-scale magnetic information storage [Science 352, 318 (2016)]. However, their potential as qubits is yet unexplored. Here we combine x-ray absorption spectroscopy and scanning tunneling microscopy (STM) with density functional theory and multiplet calculations to estimate the performance of Er and Tm on MgO(100)/Ag(100) as spin qubit candidates. For both atoms, we infer a magnetic ground state that is suitable for quantum coherent operations. By adapting the piezoelectric model of electron spin resonance (ESR)-STM [Science Advances 6, eabc5511 (2020)] to the case of lanthanide atoms, we predict a detectable signal and a higher Rabi rate compared to the systems studied up to date [S. Reale et al., accepted in Phys. Rev. B].