O 90: Spins on Surfaces at the Atomic Scale II

Time: Thursday 15:00–17:30 Location: H11

O 90.1 Thu 15:00 H11

Relativistic Orbital Effects in Hyperfine Splittings on Surfaces — •Katharina Lorena Franzke, Wolf Gero Schmidt, and Uwe Gerstmann — University of Paderborn Warburger Str. 1 33098 Paderborn

The recent combination of electron spin resonance and scanning tunneling spectroscopy (ESR-STM) provide a new platform to access single spins of atoms and molecules on surfaces [1, 2]. Characteristic hyperfine (hf) splittings due to the interaction between the electronic spin and the magnetic moments of the nuclei can be measured and compared with theoretical predictions from density functional theory (DFT). In comparison with defects in bulk material, however, the calculated data deviates considerably from the experimental values. Limited accuracy of the xc functionals or the direct influence of the electric field of the STM-tip have been discussed as possible reasons.

Large parts of the observed discrepancies however stem from a relativistic effect, the suppression of orbital quenching at surfaces. We developed a fully relativistic method that allows the calculation of this orbital contribution for complex structures [3]. For Pb ions as well as PbPc molecules on the MgO/Ag(111) substrate, this orbital part leads to additional hf splittings in the GHz range and is thereby required to achieve overall accuracy in predicting the hf interactions of single spins in 2D nanostructures.

[1] S. Baumann, et al., Science 350, 417 (2015) [2] L. Farinacci et al., Nano Letters 22, 8470 (2022) [3] K.L. Franzke, et al. J. Phys.: Conference Series. 2701, 012094 (2024)

O 90.2 Thu 15:15 H11

Electric Control of Molecular Spins on a Surface — •Paul Greule[1], Wantong Huang[1], Máté Stark[1], Kwan Ho Au-Yeung[1], Christoph Sürgers[1], Wolfgang Wernsdorfer[1], Christoph Wolf[2], and Philip Willke[1] — [1]Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul 03760, Korea

Single electronic spins hosted by atoms or molecules are candidates for future quantum technologies. To utilize them as functional building blocks in quantum information processing it is necessary to gain local control of their quantum properties. Lately, electron spin resonance combined with scanning tunnelling microscopy (ESR-STM) was demonstrated as a versatile method to access atoms and molecules on a surface [1]. For single Ti atoms it was shown that the Zeeman energy of electron spins can be tuned via the applied bias voltage in the tunnelling junction [2] constituting an atomic-scale electric field control. In our work, we present a voltage-dependent shift of the ESR frequency for two different molecular spin systems, Iron-phthalocyanine (FePc) and FePc-Fe ferrimagnet complexes. Intriguingly, we observe a strong non-linearity in the shift connected to the molecular orbital of FePc. We rationalize this theoretically by many-body interactions with the exchange bias field of the tip. Moreover, we show how the bias voltage control can be used to detune Rabi oscillations in pulsed ESR experiments. [1] Y. Chen et al., Adv. Mater. 2022, 2107534 [2] P. Kot et al., Nat Commun 14, 6612 (2023)

O 90.3 Thu 15:30 H11

Coherent control of a single Er electron spin on surface — Dasom Choi[1,2], Yaowu Liu[1,3,4], Stefano Reale[1,3], Jeongmin Oh[1,2], We-hyo Seo[1,3], Andreas Heinrich[1,2], Soo-hyun Phark[1,3], and •Fabio Donati[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Korea — [2]Physics Department, Ewha Womans University, Korea — [3]Ewha Womans University, Seoul, Korea — [4]Department of Energy, Politecnico di Milano, Milano, Italy

Electron spins on surface provide an atomic scale qubit platform for quantum information science using scanning tunneling microscopy (STM) [Phark et al. ACS Nano 17, 14144 (2023), Wang et al. Science 382, 87 (2023)]. A bottleneck of this new platform lies in the decoherence stemming from the strong interaction with the environments. Lanthanide atoms, with their highly localized 4f electrons, offer a potential solution to this issue and make them as strong candidates of single atomic spin qubits on surfaces with a high quality factor $Q = 2\Omega T_2$ (Ω = Rabi rate, T_2 = coherence time). In this talk, we present a recent advance in the qubit quality factor, achieved using erbium (Er) atoms on a MgO surface [Reale et al. Phys. Rev. B 107, 045427 (2023)]. We performed a coherent control of the Er atoms using pulsed electron spin resonance (ESR), sensed by a nearby Ti spin [Reale et al. Nat. Commun. 15, 5289 (2024)]. Notably, a single Er spin showed a fivefold improvement in the qubit quality factor compared to the prototypical Ti spin on the same substrate [Yang et al. Science 366, 509-512 (2019)]. This result represents a step towards implementing quantum processes into atomic spin qubits on surfaces.

O 90.4 Thu 15:45 H11

Spin interactions at the periphery between atomic and condensed-matter physics — •Dmitriy Borodin, Andrés Pinar Solé, Merve Ercelik, and Andreas J. Heinrich — IBS Center for Quantum Nanoscience, Seoul, South Korea

Exchange interactions are essential for the formation of chemical bonds between atoms and dictate the electronic structure of molecules. In a condensed-matter system, a manifold of exchange mechanisms coexists, ultimately defining the local magnetic structure and phase stability. In this work, we use low-temperature scanning probe microscopy to investigate the exchange mechanisms and interaction energies between atoms on surfaces and magnetic tips. By controlling the atom-tip separations on a picometer scale, we can continuously tune the interaction energies and observe changes in the local magnetic structure of the tip. Furthermore, we explore the impact of the elemental composition of magnetic tips on their exchange interactions with surface-bound atoms and identify experimental strategies to adjust the sign and strength of these interactions.

O 90.5 Thu 16:00 H11

Switching Dynamics in Fe Spin Chains: Quantum vs. Classical Behavior — •Henrik Lichtl[1], Lukas Veldman[1], Johannes Schust[1], Nicolaj Betz[1,2], Laëtitia Farinacci[1,3], Susanne Baumann[1], and Sebastian Loth[1,2] — [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 — [3]Carl-Zeiss-Stiftung Center for Quantum Photonics Jena - Stuttgart - Ulm, Germany

That quantum spins transition to classical magnets at a certain size is well-known, but the behavior of magnets at this transition point has remained elusive. Here we develop a method to measure high speed magnetization curves in a scanning tunneling microscope (STM) that reach magnetic field sweep rates up to kT/s. This allows us to measure the magnetization reversal of antiferromagnetic few-atom spin chains, constructed of Fe atoms on a Cu2N surface. By resolving the statistics of the quantum jumps between the antiferromagnetic ground states, we can clearly distinguish between classical and quantum regimes of magnetic switching. Quantum mechanical behavior quenches rapidly with increasing size such that chains of more than five atoms in length can behave as classical magnets. The ability to modulate spin systems at high speed brings techniques that were previously reserved for bulk measurements to the atomic scale and provides deep insights into the coupling mechanisms between quantum spins and their environment.

O 90.6 Thu 16:15 H11

Many-body effects in impurity spectroscopy using ESR-STM — •Christoph Wolf[1,2], Xue Zhang[3], Jose Reina-Galvez[1,2], Jan Martinek[4], and Nicolas Lorente[5,6] — [1]Center for Quan tum Nanoscience, Institute for Basic Science (IBS), Seoul, Korea — [2]Ewha Womans University, Seoul, Korea — [3]Spin-X Institute, China University of Technology, Guangzhou, China — [4]Institute of Molecular Physics, Polish Academy of Science, Poland — [5]Centro de Fisica de Materiales, CFM/MPC (CSIC-UPV/EHU), Spain — [6]6Donostia International Physics Center (DIPC), Spain

Recent advances in understanding how harmonic electric fields drive coherent spin transitions in the ESR-STM have lead to a re-evaluation of experimentally observed phenomena in the ESR-STM spectra of atomic and molecular spin adsorbed on ultrathin insulating layers. In this talk, I will show the most up to date development of our transport approach, which is based on a single-orbital Anderson impurity model attached to magnetic leads. I will focus on two aspects: first, the DC bias control of the resonance frequency of the ESR transition, which can be interpreted as an exchange bias field. This allows the full physical characterization of the adsorbate spin and the junction parameters in ESR-STM. All simulations are qualitatively and quantitatively evaluated against experimental data of individual titanium atoms and Iron(II)phthalocyanine molecules which are prototypical spin-1/2 qubits. Second, I will discuss the optimization of quantum-coherent control in the low-current regime, which shows a distinct optimum for the quantum-bit figure of merit ΩT2.

O 90.7 Thu 16:30 H11

Driving nuclear spin transitions on a single atom using STM — •Cristina Mier Gonzalez, Hester Vennema, Evert Stolte, Jinwon Lee, and Sander Otte — Delft University of Technology, 2628 CJ Delft, The Netherlands

Nuclear spins are highly isolated from its electronic environment compared to electron spins. This degree of isolation makes nuclear spins a promising platform for quantum technologies [1]. The development of ESR-STM has made possible the indirect measurement of nuclear spins on single atoms through the hyperfine interaction [2]. More recently, single-shot read-out on Ti isotopes has shown a nuclear lifetime of about 5 seconds [3].

In this work we aim to controllably address nuclear spin transitions using ESR-STM. We propose a double resonance measurement scheme to controllably drive the nuclear spin of a ^{47}Ti isotope ($I = 5/2$). Our study paves the way for coherent manipulation of single nuclear spins using STM.

[1] J. Pla et al. Nature 496, 334-338 (2013).
[2] P. Willke et al. Science 362, 336-339 (2018).
[3] E.W. Stolte et al. arXiv:2410.0870 (2024).

O 90.8 Thu 16:45 H11

Single-shot readout of an individual nuclear spin using a scanning tunnelling microscope — •Evert Stolte[1], Jinwon Lee[1], Hester Vennema[1], Rik Broekhoven[1], Esther Teng[1], Allard Katan[1], Lukas Veldman[2], Philip Willke[3], and Sander Otte[1] — [1]Department of Quantum Nanoscience, TU Delft — [2]Institute for Functional Matter and Quantum Technologies, University of Stuttgart — [3]Physikalisches Institut, Karlsruhe Institute of Technology

Nuclear spins owe their long-lived magnetic states to their excellent isolation from the environment. At the same time, a limited degree of interaction with their surroundings is necessary for reading and writing the spin state. Therefore, detailed knowledge of and control over the atomic environment of a nuclear spin is key to optimizing conditions for quantum information applications. Scanning tunnelling microscopy (STM), combined with electron spin resonance (ESR), provides atomic-scale information of individual nuclear spins via the hyperfine interaction. However, STM has thus far only sparingly been used to investigate nuclear spins in the time domain. As such, no nuclear spin lifetimes have yet been reported. Here, we demonstrate single-shot readout of an individual ^{49}Ti nuclear spin with an STM. Employing a pulsed measurement scheme, we find its lifetime to be in the order of seconds. Furthermore, we shed light on the pumping and relaxation mechanisms of the nuclear spin by investigating its response to both ESR driving and tunnelling current. These findings give an atomic-scale insight into the nature of nuclear spin relaxation and are relevant for the development of atomically assembled qubit platforms.

O 90.9 Thu 17:00 H11

Spin Excitations of High-Spin Fe(II) in Metal-Organic Chains on Metal and Superconductor — •Jung-Ching Liu[1,2], Chao Li[2], Outhmane Chahib[2], Xing Wang[3], Simon Rothenbühler[4], Robert Häner[4], Silvio Decurtins[4], Ulrich Aschauer[5], Shi-Xia Liu[4], Ernst Meyer[2], and Rémy Pawlak[2] — [1]Department of Physics, Technical University of Munich, James-Franck-Str.1, 85748 Garching, Germany — [2]Department of Physics, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland — [3]Paul Scherrer Institut, Forschungsstrasse 111, 5232 Villigen PSI, Switzerland — [4]Department of Chemistry and Biochemistry, University of Bern, Freiestrasse 3, 3012 Bern, Switzerland — [5]Department of Chemistry and Physics of Materials, University of Salzburg, Jakob-Haringer-Strasse 2A, 5020 Salzburg, Austria

Magnetic anisotropy induced by the substrate plays an important role in many-body interactions in metal-organic frameworks at surfaces. Investigated by STM at 1K, we present the study of magnetic signature at Fe atoms with high spin-state (S=2) using PTO as ligands. On Ag(111), we found long-range Fe-Fe coupling in addition to low-energy spin-flip excitations. Despite of the identical chain structure, such long-range superexchange through PTO is not observed on tunneling spectra on Pb(111) superconductor. We ascribe this distinct spin-spin coupling behavior to the depletion of electronic states around the Fermi level on Pb(111) as compare to Ag(111). We believe our study provides a route for fundamental studies in spin-spin and spin-substrate interactions with different lattice structures.

O 90.10 Thu 17:15 H11

Control of the Landau-Zener Gap in Atomic Structures — •Piotr Kot[1,2], Yaowu Lei[1,2], Valeria Sheina[1,2], We-hyo Seo[1,2], Andreas Heinrich[1,2], and Soo-hyon Phark[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, South Korea — [2]Department of Physics, Ewha Womans University, Seoul, South Korea

Avoided crossings in quantum systems have been shown to be a useful tool for the study of qubits and the actualization of faster qubit operations. This is done by taking advantage of Landau-Zener transitions, allowing one to measure quantum interferometric effects. However, implementing these techniques in on surface atoms has not yet been demonstrated experimentally, and is a crucial next step for studying this new class of spin qubits. Here we present preliminary steps towards studying quantum interferometric effects in Ti dimers using electron spin resonance scanning tunnelling microscopy. Firstly, we have fine-tuned the Landau-Zener gap in our dimers by studying the dipole and exchange interaction between the atoms. We have found that only several dimers have the correct gap magnitude for our experimental conditions. Secondly, we have found that by using three atom structures we are able to position the avoided crossing within our range of interest. Experiments in the near future will include measuring Landau-Zener transitions, Stueckelberg oscillations and quantum interferometry maps.