Location: R2

O 44: Mini-Symposium: Manipulation and control of spins on functional surfaces II

Time: Tuesday 13:30-15:30

Invited Talk O 44.1 Tue 13:30 R2 Atomic-scale spin sensing with a single molecule at the apex of a STM — •LAURENT LIMOT — Université de Strasbourg, CNRS, IPCMS, Strasbourg, France

The decoration of metal probe-tips by a molecule intentionally picked up from a surface has proven to be a powerful method to improve the measurement capabilities of a scanning tunneling microscope (STM). The degrees of freedom of the molecule introduce tip-surface interactions across the vacuum gap that are usually absent when using a metallic apex. These interactions can endow STM with an enhanced sub-molecular resolution and provide new chemical insight.

The success of this approach opens the prospect of introducing spin sensitivity through the tip functionalization by a magnetic molecule. We show here that it is possible to use metallocene-terminated tips to monitor surface magnetism through the inelastic component of the tunneling current, which provides an electrical access to the metallocene spin states. When the tip is 100 picometers away from point contact, the exchange interaction between the tip and a magnetic sample changes the metallocene spin states. This detection scheme can then be used to simultaneously probe the sample exchange field and spin polarization with atomic-scale resolution.

O 44.2 Tue 14:00 R2 Free coherent evolution of a coupled atomic spin system initialized by electron scattering — LUKAS M. VELDMAN¹, •LAETITIA FARINACCI¹, RASA REJALI¹, RIK BROEKHOVEN¹, JEREMIE GOBEIL¹, DAVID COFFEY¹, MARKUS TERNES^{2,3}, and ALEXANDER F. OTTE¹ — ¹Delft University of Technology, The Netherlands — ²RWTH Aachen University, Germany — ³Peter-Grünberg-Institute, Jülich, Germany

Observing the free evolution of a coupled spin system is an essential step towards studying collective quantum spin dynamics, as well as gaining insight into the fundamental mechanisms leading to spin excitation. Here, we combine pump-probe and ESR techniques with STM to study the free evolution of a single atomic spin depending on its level of entanglement with a second one. We build TiH dimers on MgO/Ag(100) in which the two spins are inherently detuned. We then make use of the magnetic interaction with the STM tip to tune the level of entanglement between the two spins: using ESR, we characterize the energy diagram of the dimer and identify the tip height at which both spin precess at the same frequency. Subsequently, we use a pump-probe scheme to, first, initialize the system via an electron induced spin excitation and, second, study the free evolution of the spin under the tip. We show that only when the two spins entangle, the excitation is swapped back and forth at a frequency that is given by their coupling strength. These results provide insight into the locality of electron-spin scattering: only the spin directly underneath the tip is affected, irrespective of its global quantum state.

Invited Talk O 44.3 Tue 14:15 R2 Quantum sensing and operation of single molecules on the surface — •Xue Zhang — Center for Quantum Nanoscience, Insti-

tute for Basic Science (IBS), Seoul 03760, Republic of Korea — Ewha Womans University, Seoul 03760, Republic of Korea

Scaling down information devices to atom-scale has brought the interest of using individual spins as basic unit for data storage. Scanning tunneling microscopy (STM) combined with fast electric pulse and electron spin resonance (ESR) technique has been proved a powerful tool to access and coherently control individual spins in atomic scale.

Here, we investigated the spin properties and dynamics of artificially built Fe-tetracyanoethylene (TCNE) complexes by using electronic spin pump-probe spectroscopy. Further, we spatially imaged the location of the spin center within the complex [J. Phys. Chem. Lett., 2020, 11, 14, 5618]. This work highlights that the combination of STM with electronic spin relaxometry can provide highly valuable clues for investigating magnetic metal-organic nanostructures. While current ESR-STM studies have focused on atom spins, we achieved driving ESR on single molecules (metal phthalocyanine) on a bilayer magnesium oxide (MgO) surface atop Ag(100) [under review]. We probed and differentiated the exchange and dipole interaction between molecular dimers. This work demonstrates the feasibility of employing single molecules in atom-scale quantum control studies and shed light on intriguing magnetic interactions between molecular spins, which is crucial for developing molecule-based spintronic devices.

Combining the sub-atomic resolution of scanning tunneling microscopy (STM) with the spectral resolution of electron-paramagnetic resonance (EPR) allows for sensitively probing magnetic interactions of single atoms on a surface [1]. However, the experimental requirements for driving the EPR transitions are still under debate. In-depth understanding of the EPR-STM driving is mandatory to explore novel material systems and optimize the sensitivity of this technique. Here, we acquire and model EPR spectra of single Fe and hydrogenated Ti atoms on bilayer MgO on Ag [2]. We investigate the impact of radiofrequency excitation strength and tunneling parameters on the EPR signal and find strong evidence for a piezoelectric coupling mechanism [3]. In this mechanism, the surface atom oscillates at radiofrequencies in the inhomogeneous tip magnetic field. Based on density functional theory and atomic-multiplet calculations, we reveal different driving mechanisms for single Fe and hydrogenated Ti atoms on the surface. Specifically, transverse magnetic field gradients drive the spin-1/2 hydrogenated Ti, whereas longitudinal magnetic field gradients drive the spin-2 Fe. [1] S. Baumann et al., Science 350 (2015); [2] T. S. Seifert et al., PRR 2 (2020); [3] T. S. Seifert et al., Sci. Adv. 6 (2020)

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