O 3: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies I

Spins on surfaces represent a prospering field fueled by advancements in scanning probe microscopies and quantum impurity theory. This research spans three key areas: spins on superconductors revealing subgap states, spins on normal metals showcasing unexpected Kondo effects, and spins on thin insulating films enabling coherent spin control and qubit gate operations. These phenomena, united by fundamental principles, present intriguing challenges such as extending coherence times and establishing coherent coupling between different spin systems.

Organizers: Christian Ast (MPI Stuttgart), Susanne Baumann, Sebastian Loth (U Stuttgart)

Time: Monday 10:30–12:15 Location: MA 004

Topical Talk O 3.1 Mon 10:30 MA 004

Magnetic molecules on superconductors: from sensing experiments to novel molecular candidates for quantum platforms — •Giulia Serrano — Department of Industrial Engineering, University of Florence, florence, Italy

The interaction between magnetic molecules and superconductors (SC) has unveiled intriguing phenomena with implications for spintronics and quantum technologies. A monolayer of Single Molecule Magnets, characterized by magnetic hysteresis, has demonstrated a remarkable sensitivity to the SC transition. This sensitivity influences their hysteresis and triggers the activation of the resonant magnetization regime [1],[2].

At the single molecule level, the emergence of localized magnetic states, resulting from the interplay between the spin and the SC surface, holds the potential to pave the way for more sophisticated quantum platforms, such as those involving topologically protected qubits. In this context, innovative molecular systems are proposed, wherein ligands mediate spin-spin interactions. Their preliminary exploration on surfaces is presented [3],[4].

[1] Serrano et al., Nat. Mater., 19, 546, 2020. [2] Serrano et al., Nat. Commun., 13, 3838, 2022. [3] Santanni et al., JACS Au, 3, 1250, 2023. [4] Ranieri et al., Chem. Sci., 14, 61, 2023.

O 3.2 Mon 11:00 MA 004

Magnetic field dependence of Yu-Shiba-Rusinov (YSR) excitations — •Niels P.E. van Mullekom[1], Benjamin Verlhac[1], Werner M.J. van Weerdenburg[1], Hermann Osterhage[1], Manuel Steinbrecher[1], Katharina J. Franke[2], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University Nijmegen, the Netherlands — [2]Fachbereich Physik, Freie Universität Berlin, Germany.

Yu-Shiba-Rusinov (YSR) states arise from the exchange coupling between a local spin and a superconductor and can be characterized by resonances inside the gap superconducting gap. From the in-gap excitations alone, the interplay of this exchange interaction with other energy scales, as well as the role of higher spin degrees of freedom is not clear. Magnetic field dependent characterization could clarify this, but most studies of YSR states to date have been limited to bulk superconductors, which easily quench in the presence of modest magnetic fields.

Utilizing the enhanced critical fields of a thin superconducting film we characterize the magnetic phase diagram of a molecule on the surface using high resolution milliKelvin scanning tunneling microscopy and spectroscopy. We observe nontrivial changes in the YSR excitations, that go beyond the trends that are expected in a spin 1/2 picture. We relate these changes to the various properties of the molecule, such as the role of multiple channels and magnetic anisotropy. These results provide an in-depth and detailed approach to understand the role of high spin states in the YSR excitations.

O 3.3 Mon 11:15 MA 004

Dynamics of individual Yu-Shiba-Rusinov states measured with stochastic resonance spectroscopy — •Nicolaj Betz[1], Vivek Krishnakumar Rajathilakam[1], Susan Coppersmith[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]School of Physics, University of New South Wales, Sydney, Australia.

Coupling a spin to non-ohmic environments such as a superconductor can lead to new behavior with interesting dynamics. Thus far, the dynamics of emerging states like Yu-Shiba-Rusinov (YSR) states at individual atoms or molecules have mostly been investigated through tunneling rates [1,2]. Here, instead, we show a measurement that uses the synchronization of the state occupation with a frequency-dependent drive voltage to measure the dynamics of the tunneling through YSR states themselves. This technique relies on stochastic resonance which was previously used to measure spin dynamics [3]. We use a scanning tunneling microscope (STM) to measure the dynamics of YSR states of Fe atoms adsorbed on the oxygen-reconstructed surface of V(100) and find YSR state lifetimes in the picosecond range. This work provides a new level of insights into the dynamics of YSR states and can lead to a better understanding of relaxation mechanisms in spin-superconductor structures.

[1] M. Ruby et al., PRL 115, 087001 (2015).
[2] H. Huang et al., Nat. Phys. 16, 1227-1231 (2020).
[3] M. Hänze et al. Sci. Adv. 7, eabg2616 (2021).

O 3.4 Mon 11:30 MA 004

Electron transport in magnetic atomic and molecular junctions between superconducting leads — •Nicolas Lorente[1,2,3], Divya Jyoti[2,3], Cristina Mier[2,3], and Deung-Jang Choi[2,3] — [1]Centro de Física de Materiales (CSIC-EHU), San Sebastian, Spain — [2]Materials Physics Center, San Sebastian, Spain — [3]Donostia International Physics Center, San Sebastian, Spain

We present experiments and theory using self-consistent Bogoliubov-de Gennes equations implemented with Green's function for wide-band s-wave superconductors. We show the effect of magnetic junctions and multiple Andreev reflection when there are localized magnetic moments in the junction. The theoretical results agree with experiments performed on Nickelocene molecules trapped between a superconducting tip and a superconducting surface. Adding Fe atoms to the junctions enhances the magnetic scattering which can be easily modeled with the theory. Experimentally, the Fe atoms can be manipulated to be between molecule and interface or to be subsurface, with very different signature in the differential conductance. Self-consistency does not alter the results except when studying the evolution of the order parameter, which has bearings in the prediction of normal and topological quantum phase transitions.

O 3.5 Mon 11:45 MA 004

Magnetic bound states of iron clusters on a superconductor — •Ankur Das[1], Silas Amann[1], Nóra Kucska[2], András Lászlóffy[2], Nicolas Néel[1], Balázs Újfalussy[2], Levente Rózsa[2,3], Krisztián Palotás[2,3,4], and Jörg Kröger[1] — [1]Institut für Physik, Technische Universität Ilmenau, D-98693 Ilmenau, Germany — [2]Department of Theoretical Soild-State Physics, HUN-REN Wigner Research Centre for Physics, Institute for Solid State Physics and Optics, H-1525 Budapest, Hungary — [3]Department of Theoretical Physics, Institute of Physics, Budapest University of Technology and Economics, H-1111 Budapest, Hungary — [4]HUN-REN-SZTE Reaction Kinetics and Surface Chemistry Research Group, University of Szeged, H-6720 Szeged, Hungary

The magnetic exchange interaction of Fe_n (n=1-3) clusters with superconducting Pb(111) is probed by scanning tunneling spectroscopy of Yu-Shiba-Rusinov states. The Yu-Shiba-Rusinov resonances are shifted from the coherence peaks in the Fe monomer spectrum towards the Fermi energy in the Fe dimer spectrum. Unexpectedly, the linear Fe trimer does not follow this trend, as it exhibits an almost identical spectrum to the single Fe atom. Kinked Fe trimers in contrast show strong Yu-Shiba-Rusinov resonances well within the Bardeen-CooperSchrieffer energy gap of the substrate. First-principles simulations of the Yu-Shiba-Rusinov states reveal which adsorption geometries and magnetic structures of the clusters can reproduce the experimental spectra most accurately.Funding by the Deutsche Forschungsgemeinschaft (KR 2912/18-1) is acknowledged.

O 3.6 Mon 12:00 MA 004

Ligand-mediated Yu-Shiba-Rusinov states of a S=1/2 molecule — •Lukas Arnhold[1], Matteo Briganti[2], Andrea Sorrentino[3], Nicolaj Betz[1], Lorenzo Poggini[4], Jaisa Fernandez Soarez[5], Luana C. de Camargo[5], Giulia Serrano[3], Matteo Mannini[2], Federico Totti[2], Roberta Sessoli[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Department of Chemistry Ugo Schiff, University of Florence, Italy — [3]Department of Industrial Engineering, University of Florence, Italy — [4]Institute of Chemistry of OrganoMetallic Compounds - CNR, Florence, Italy — [5]Department of Chemistry, Federal University of Paraná, Brazil

We couple ((*8-cyclooctatetraene)(*5-cyclopentadienyl)titanium) (CpTicot) molecules to superconducting Pb nanoislands on Si(111) in a scanning tunneling microscope (STM). This molecular magnet features a well isolated S=1 /2 ground state and is a prime candidate for molecular qubit applications due to exceptional spin coherence times in frozen solution [1]. On Pb nanoislands it exhibits a single YSR state stemming from a singly occupied molecular orbital (SOMO). We find that the molecule's spin interacts with the Pb surface via the cyclooctatetraene ligand making it possible to link the electronic properties of the molecule quantitatively to the in-gap YSR states both in theory and experiment. This experiment gives insights into chemical design priciples for targeted applications of YSR states.

[1] de Camargo, L., et al. Angewandte Chemie. 60, Issue 5, 2588-2593