Surface Science
Room 209 CDE W - Session SS-ThM
Surface Electrical, Magnetic, and Optical Properties

Moderators: Melanie Müller, Fritz Haber Institute of the Max Planck Society, **Shadi Fatayer**, King Abdullah University of Science and Technology

8:00am **SS-ThM-1 Storing and Processing Information in the Magnetic Quantum States of Single Surface Adsorbed Atoms***, **Harald Brune**,* Swiss Federal Institute of Technology Lausanne, Switzerland **INVITED**

The magnetic properties of single surface adsorbed atoms became one of the core interests in surface and nanoscience in 2003, where single Co atoms on Pt were reported to have 200 times the magnetic anisotropy energy of bulk Co [1]. Even 1000 times this energy was reached for single Co atoms on thin MgO films [2]. In a classical picture, this suggests that these single atoms should be rather stable magnets. However, despite numerous efforts, the magnetic quantum states of all investigated single surface adsorbed transition metal atoms had very short magnetic relaxation times, below 1 μs.

Immediately after changing from 3*d* elements to rare-earth atoms, a few adsorbate/substrate combinations could be identified, where the magnetization vector of a single atom is indeed stable over hours in the absence of an external magnetic field [3,4]. Therefore, these systems are single atom magnets and enable magnetic information storage in the smallest unit of matter. We will give an overview over the present adsorbate/substrate systems exhibiting single atom magnet behavior [3 – 10] and explain the essential ingredients for this surprising stability of single spin systems that are exposed to numerous perturbations from the environment. These atoms can be placed very close and still individually be addressed, conceptually enabling information storage at densities by 3 orders of magnitude larger than presently used devices.

Now the fundamental research field turns its attention to quantum coherent spin operations in single surface adsorbed atoms. If they have long enough coherence times with respect to the time it takes to perform a single quantum spin operation, these would be single atom quantum bits. The requirements for long coherence times of the magnetic quantum states are quite different from the ones of magnetic relaxation times. We will illustrate this with a few examples and point out single rare-earth atom systems that lend themselves already now as quantum repeaters in telecommunication [11], creating hope that single atom qubits may indeed become reality [12].

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8:30am **SS-ThM-3 Nonadiabatic Dynamics Simulations of Carbon Atom Scattering from Au(111)***, **Alexander Kandratsenka**,* MPInat, Germany

Measurements of energy loss spectra of carbon atom scattered off Au(111) surface conducted recently at Dalian Coherent Light Source facility suggest involvement of nonadiabatic effects. In order to construct a theoretical model describing that we are going to use the Independent-Electron Surface Hopping (IESH) approach, where the interaction of an incident atom (or molecule) with the surface induces the overlap of atomic and metallic orbitals facilitating energy exchange between projectile's nuclear degrees of freedom and ehp of a metal. The first step here is to produce full-dimensional potential energy surfaces for each single-electron state relevant for the dynamics. In case of C atom, these are triplet (ground state) and singlet spin states of a neutral C, and quartet and doublet states of C anion. The second step is to perform nonadiabatic dynamics simulations to study the influence of Intersystem Crossing regions on the carbon atom energy loss spectra.

8:45am **SS-ThM-4 Oxidation of Ni-Based Superalloys: Closing the Gap from Adsorption to Microstructure***, William Blades,* Juniata College*; Keithen Orson,* University of Virginia, USA*; Juran Niu, Alexei Zakharov,* MAX IV Laboratory, Sweden*; Jerzy Sadowski,* Brookhaven National Laboratory*; **Petra Reinke**,* University of Virginia, USA

Ni-based superalloys are coveted for their corrosion resistance and formation of highly inert passive layers which limit degradation in a wide range of environments. Our work probes the oxidation process from the initial oxygen adsorption on a pristine alloy to the nanometer scale oxide layer using scanning tunneling microscopy and spectroscopy (STM/STS), and electron spectroscopies including X-ray photoelectron spectroscopy (XPS), and X-ray photoemission electron spectroscopy (XPEEM). We bridge the gap between single atom alloy to the complex solid solution surface, and realistic microstructure.

The Ni-Cr system has become a widely used model system for the study of oxidation, and passivity. It reflects the competition between Ni and Cr oxidation, the interplay between thermodynamic preference for chromia formation and kinetic limitations imposed by reactant transport in the alloy. The main factors which control the oxidation are alloy composition, temperature and crystallographic orientation. We will discuss the interplay between these factors on the oxide nucleation and growth between 200°C to 600°C with 5at% to 22at%Cr.

The delayed nucleation of NiO on Ni(100) is lifted on Ni-15Cr(100) leading to the rapid growth and step edge reconstruction with NiO. The chromia nuclei remain spherical due to their lack of epitaxial preference, although local segregation of Cr islands and a Cr(100)p(2´2) reconstruction is observed. Distinct variations in nucleation rate occur as a function of crystallographic orientation on Ni-15Cr(111) and several higher index surfaces on polycrystalline alloys. The direct observation of chromia nucleation on Ni-22Cr and Ni-22Cr-Mo with XPEEM opens the window to understanding the role of Mo, whose addition leads to high quality, chromia dominated, protective oxide layers. Adding Mo, or W modulates the surface chemistry towards chromia formation, and switches the growth to a layer-by-layer mode. Both, the barrier towards chromia formation, and the chromia-alloy interfacial energies are modified in favor of a dense protective layer.

A model which represents the alloy surface as a random solid solution will be used to capture adsorbate induced segregation as the initial step leading from adsorption towards oxide nucleation. The distribution of Cr in the surface is captured using fractal dimensions, percolation models and distribution functions combined with a systematic variation of diffusion constants for Cr. The outcome of the calculations is compared to nuclei densities and distributions from experiment.

9:00am **SS-ThM-5 Induced Fit of Adsorbates Inside Nanometer-Sized on-Surface Quantum Confinements***, **Aisha Ahsan**,* University of Basel, Switzerland*; Thomas Jung,* Paul Scherrer Institute, Switzerland*; Lutz Gade,* University of Heidelberg, Germany*; Luiza Buimaga Iarinca,* National Institute for Research and Development of Isotopic and Molecular Technologies, 67-103 Donat, 400293 Cluj-Napoca, Romania

Condensation processes are important in physics and chemistry especially when Host-guest buildings provide ideal systems to study site-specific physical and chemical effects. In this scenario conformationally flexible molecules, in contrast, may behave characteristically different due to the possibility to flex upon adsorption and upon compression inside confinements. As objects of study we have chosen a series of non-planar, conformationally flexible cyclo-alkanes (c-alkanes) to study their site-specific condensation in the confinement provided by 1.6 nm sized pores in a surface supported coordination network. They may be accompanied by conformational adjustments representing induced fit packing patterns. We report that the symmetry of small clusters formed upon condensation, their registry with the substrate, their lateral packing as well as their adsorption height is characteristically modified by the packing of cycloalkanes in confinements. While cyclopentane and cycloheptane display cooperativity upon filling of the hosting pores, cyclooctane and to a lesser degree cyclohexane diffusively re-distribute to more favored adsorption sites. The dynamic behavior of cyclooctane is surprising at 5K given the cycloalkane melting point above 0°C. The internal reorientation of molecules is reminiscent of the induced fit concept invoked for enzyme/substrate interactions and is expected to be a general phenomenon for such inclusion processes, both on surfaces and in porous solids. The ability to address these individually and to investigate local properties in a one-by-one fashion is a new opportunity. It may facilitate the rationally driven development of functional materials, catalysts, in which such molecule-host interactions at interfaces play a crucial role.

9:15am **SS-ThM-6 Chemically Interrogating N-Heterocyclic Carbenes at the Single-Molecule Level Using Tip-Enhanced Raman Spectroscopy*, Nan Jiang,*** University of Illinois - Chicago

N-heterocyclic carbenes (NHCs) have been established as powerful modifiers to functionalize metal surfaces for a wide variety of energy and nanoelectronic applications. To fundamentally understand and harness NHC modification, it is essential to identify suitable methods to interrogate NHC surface chemistry at the spatial limit. Here, we demonstrate tip-enhanced Raman spectroscopy (TERS) as a promising tool for chemically probing the surface properties of NHCs at the single-molecule scale. We show that with subnanometer resolution, TERS measurements are capable of not only unambiguously identifying the chemical structure of individual NHCs by their vibrational fingerprints but also definitively determining the binding mode of NHCs on metal surfaces. In particular, by investigating low-temperature NHC adsorption on Ag(111), our TERS studies provide insights into the temperature dependence of the adsorption properties of NHCs. Furthermore, we investigate the mobility of a model NHC on Ag(111). Two distinct molecular behaviors are observed, depending on substrate preparation. Room-temperature deposition leads to diffusing NHC–Ag adatom complexes exhibiting a ballbot-like motion, chemically identified by TERS through their spectroscopic fingerprint. By contrast, NHCs deposited at low temperature are stabilized on Ag(111) as isolated single molecules directly bound to the substrate. Significantly, a desorption/readsorption scenario is suggested for the displacement of NHCs by moving otherwise immobile single NHCs deposited at low temperature via STM manipulation, with their trajectory traced to atomic precision. This work suggests the potential of single-molecule vibrational spectroscopy for investigations of NHC surface modification at the most fundamental level.

9:30am **SS-ThM-7 Atomic-Scale Spectroscopy of Ultrafast Charge Order Dynamics in Charge-Density Wave Materials*, Sebastian Loth,*** University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Germany **INVITED**

In materials with mobile electrons, electron-phonon coupling and electron-electron interaction can cause the emergence of charge-ordered phases, such as charge-density waves or Mott insula-tors. These ordered electronic states feature collective excitations that are absent in conventional metals but are fundamental to understanding correlated electron physics. By combining scanning tunneling microscopy with terahertz excitation (THz-STM) [1,2], we achieve simultaneous atomic spatial and femtosecond temporal resolution to directly visualize these dynamics at their intrinsic length and time scales.The extreme field enhancement at the STM tip apex produces localized THz fields reaching MV/cm [3]. This enables localized excitation of surfaces by the electric field of the THz lightwave through THz-induced Coulomb forces [4] and ultrafast screening currents. We apply this technique to two prototypical CDW systems: the incommensurate CDW in 2H-$NbSe_2$ and the commensurate CDW in 1T-TaS_2. In $NbSe_2$, we observe collective phase excitations at sub-THz frequencies that originate from atomic pinning sites, revealing how disorder dictates local dynamics and creates heterogeneity in the response [5]. In contrast, TaS_2 exhibits rapid reconfigurations within individual domains that show layer stacking dependence.These measurements demonstrate how ultrafast THz excitation at surfaces can manipulate electronic order in quantum materials with extreme spatial precision. This approach provides insights into the atomic-scale mechanisms governing CDW pinning. The ability to resolve these fluctuations in real space at the scale of individual impurities provides a new route to unraveling the electronic dynamics of disordered correlated materials. References:[1] Cocker., T. L., et al. *Nat. Photon.* 7 620 (2013). [2] Cocker., T. L., et al. *Nature* 539 7628 (2016). [3] Abdo, M., et al. *ACS Photonics* 8 702 (2021). [4] Sheng, S., et al. *Phys. Rev. Lett.* 129 043001 (2022).[5] Sheng, S., et al. *Nat. Phys.* 20 1603 (2024).

11:00am **SS-ThM-13 Measuring Properties of Single Defects, Dopants and Quantum Dots with nm Spatial Resolution*, Peter Grutter,*** McGill University, Canada **INVITED**

Semiconductor interfaces often have isolated trap states which modify electronic properties. We have developed a framework to quantitatively describe a metal-insulator semiconductor (MIS) device formed out of a metallic AFM tip, vacuum gap, and semiconducting sample. This framework allows the measurement of local dopant concentration, bandgap and band bending timescales with nm scale resolution of different types of defects on semiconductors such as Si, 2D $MoSe_2$ and pentacene monolayers [1].

With this method, we have characterize individual defects at the Si-SiOx interface. We show that surface charge equilibration timescales, which range from 1−150 ns, increase significantly around interfacial states [2]. We conclude that dielectric loss under time-varying gate biases at MHz and sub-MHz frequencies in metal-insulator-semiconductor capacitor device architectures is highly spatially heterogeneous over nm length scales. We have also analyzed two-state fluctuations localized at these interfacial traps, exhibiting bias-dependent rates and amplitudes. When measured as an ensemble, these observed defects have a 1/f power spectral trend at low frequencies [3]. Low-frequency noise due to two level fluctuations inhibits the reliability and performance of nanoscale semiconductor devices, and challenges the scaling of emerging spin based quantum sensors and computers. The presented method and insights provide a more detailed understanding of the origins of 1/f noise in silicon-based classical and quantum devices, and could be used to develop processing techniques to reduce two-state fluctuations associated with defects.

Force detection with single electron sensitivity can be used to perform localized electron energy level spectroscopy on semiconductor quantum dots, individual ferrocene molecules and atomically precisely positioned dopant atoms in Si. Single electron force spectroscopy allows the measurement of Coulomb blockade and eigen state energy levels, shell structure, excited state energies, coupling strength to electrodes, molecular vibrations, reorganization energies, electron-nuclear coupling (Franck-Condon blockade), stability diagrams (i.e. coupling between qdots) and double dot coherence time (for a recent review see [4]). I will describe the challenges and progress towards applying this single electron force spectroscopy technique to atomically precisely positioned quantum dots in Si.

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11:30am **SS-ThM-15 Activating Elastic Conformation of a Single Molecule via Qplus AFM Tip*, Markus Zirnheld,*** *A.M. Shashika D. Wijerathna, Yuan Zhang,* Old Dominion University

Mechanical properties of molecules adsorbed on material surfaces are increasingly vital for the applications of molecular thin films. In this study, we induce molecule conformational change on a single molecule through the application of mechanical load and quantify the associated force and energy required via a combination of a low temperature (~ 5.4 K) ultra-high vacuum (~10^{-10} Torr) Scanning Tunneling Microscope (STM) and Qplus Atomic Force Microscope (Q+AFM). The molecule under investigation, TBrPP-Co (II) (a cobalt porphyrin), deposited on an atomically clean gold substrate, typically has two pyrrolic units (resembling pentagon rings) tilted upward and the other two downward. An atomically sharp Pt/Ir tip of the STM/Q+AFM, which vibrates with a high frequency (~ 30kHz), is employed to run over a single TBrPP-Co(II) molecule at different heights with 0.1 Å as decrements and meanwhile to record tip-molecule interaction strength in the form of tip frequency change. When the tip approaches the threshold distance to the molecule, the mechanical load by the tip becomes large enough to trigger the elastic conformation of the molecule and cause pyrrolic units to flip their orientation in the opposite direction. Due to the sensitive nature of tip-molecule interaction, the pyrrolic units flipping can be directly visualized by STM, where upward-tilted pyrrolic units appeared as two bright protrusions, contrasting with the appearance of downward-tilted pyrrolic units. By processing frequency change, we obtain a three-dimensional mechanical force and potential maps for the single molecule TBrPP-Co(II) with the resolution of angstrom level. Our results indicate that a potential barrier of ~ 49 meV is needed to activate the elastic conformational switch responsible for inducing pyrrolic units flipping of a single TBrPP-Co(II) molecule.

KEYWORDS

single molecular switch, mechanical load, qplus atomic force microscope, energy barrier, elastic conformational change, porphyrin molecule

11:45am **SS-ThM-16 Local Superconductor-to-Semiconductor Phase Transition in WS2 Controlled by STM Tip***, **TeYu Chien**,* University of Wyoming

Transition metal dichalcogenides (TMDs) are a unique class of materials that often host electronic correlation and strong spin orbital coupling. TMDs are two-dimensional (2D) layered materials with van der Waals (vdW) force between the layers allowing various stacking structures. Thus, TMDs often have polymorphic crystal structures, which exhibit drastically different physical properties. In WS_2, the most stable 2H phase is a topologically trivial semiconductor, while the metastable 2M phase is superconducting (SC) with critical temperature of 8.8 K. The 2M phase is also reported to be possible topological SC. A zero-bias peak has been reported inside magnetic vortices via scanning tunneling microscopy (STM) measurements and is considered to be a candidate of the Majorana Zero Modes (MZMs). It also has been reported that the metastable 2M phase WS_2 can be converted into 2H phase by heating. Thus, it is interesting to explore the possibility of creating topological SC anti-dot in nm scale to host the potential MZMs. In this study, we demonstrate a precise control of 2M to 2H phase transition in WS_2 using a STM tip “current pulsing” method. The resulting phase transition areas are notably sharply hexagonal following the 2H lattice orientation and can range from 30-350 nm in diameter. The effects of the electric field and tunneling current on the conversion will be discussed to provide insights of the conversion mechanism.

Funding acknowledgement: NSF OSI-2228841

12:00pm **SS-ThM-17 Multimodal Tool Combining Multichannel HREELS and ARPES/XPS to Study Electron-Boson Coupling***, **Takahiro Hashimoto,** TImo Wåtjen,* Scienta Omicron AB, Sweden*; Xin Zhang, Andrew Yost, Daniel Beaton,* Scienta Omicron, Inc.*; Harald Ibach, Stefan Tautz, François Bocquet,* Forschungszentrum Jülich GmbH, Germany

Electrons in quantum materials couple to bosonic excitations, such as phonons and magnons, making it essential to characterize both these excitations and the electronic band dispersion. Multimodal characterization, where a single sample is analyzed using multiple complementary techniques, offers a powerful approach to uncovering the interplay between various excitations and electronic structure. High-resolution electron energy loss spectroscopy (HREELS) is a technique for observing surface excitations including phonons, magnons, plasmons, excitons, and vibrational modes. Multimodal characterization has been difficult with a traditional single channel HREELS instrument because it requires its own detector, the measurements are time consuming, and angular resolution is limited. To improve the efficiency of HREELS measurements and to realize multimodal measurement with photoemission spectroscopy, we developed a solution by combining a monochromatic collimated electron source and a hemispherical electron analyser, commonly used for ARPES and XPS. The multichannel 2D detector of the analyser simultaneously measures hundreds of channels in both the energy and angular directions, and the measurements are orders of magnitude faster than the single channel setups. By adding a light source for photoemission spectroscopy, this setup becomes a multimodal characterization tool that combines state-of-the-art HREELS and ARPES/XPS using the same electron analyser. It allows to study the interplay between various surface excitations and electronic properties including electron-phonon coupling and electron-magnon coupling. Also, recent examples of HREELS measurements, including anisotropic exciton dispersion and topological phonons, are discussed to showcase the power of multichannel HREELS to observe novel excitations.

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