O 84: Gerhard Ertl Young Investigator Award Competition

Time: Thursday 10:30–13:00 Location: H24

O 84.1 Thu 10:30 H24

CO cryo-sorption on a FeNC catalyst as a spectroscopic probe of the active site density — •Benedikt P. Klein[1,3], Beomgyun Jeong[1], Hafiz Ghulam Abbas[2], Geunsu Bae[2], Adith R. Velmurugan[2], Chang Hyuck Choi[4], Geonhwa Kim[5], Dongwoo Kim[5], Ki-jeong Kim[5], Byeong Jun Cha[6], Young Dok Kim[6], Frederic Jaouen[7], Reinhard J. Maurer[3], and Stefan Ringe[2] — [1]Korea Basic Science Institute, Daejeon, ROK — [2]Korea University, Seoul, ROK — [3]University of Warwick, Coventry, UK — [4]POSTECH, Pohang, ROK — [5]PAL, Pohang, ROK — [6]Sungkyukwan University, Suwon, ROK — [7]University of Montpellier, France

To quantify the number of active sites in a catalyst is a crucial necessity for the evaluation of its performance. A possible quantification approach is the adsorption of a gas molecule to the catalytically active sites as a probe. If the adsorbed molecules can be reliably detected, their surface density can be determined by using spectroscopic techniques and the active site density of the catalyst can be obtained. For this approach it is necessary to identify the probe molecules according to their spectroscopic features, a task much helped by the computational simulation of spectra. In this the NAP-XPS and NAP-NEXAFS study, we present direct spectroscopic evidence for the adsorption of CO on an iron-nitrogen-carbon (FeNC) catalyst. The well defined spectroscopic features enable us to determine the active site density using by experimental data combined with state-of-the-art DFT based spectroscopy simulation.

O 84.2 Thu 11:00 H24

Nano-optical imaging of grating-coupled waveguide modes in 3R-MoS$_2$ — •Fabian Mooshammer[1], Xinyi Xu[2], Chiara Trovatello[2], P. James Schuck[2], and D. N. Basov[3] — [1]Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, Regensburg, Germany — [2]Department of Mechanical Engineering, Columbia University, New York, USA — [3]Department of Physics, Columbia University, New York, USA

Waveguide modes in thin slabs of van der Waals crystals serve as reliable probes of the anisotropic dielectric response of the material and also hold great potential for the implementation of on-chip optical elements. Here, we show that near-field imaging can resolve the propagation patterns of waveguide modes, which encode the birefringence of transition metal dichalcogenides. We use a laser patterning procedure to write grating structures into 3R-MoS$_2$ crystals with grooves as sharp as 250 nm. Spatial maps of the scattered electric field in the vicinity of the gratings reveal a directional, polarization-splitting outcoupling of waveguide modes, which is in line with finite element simulations. Experiments over a range of near-infrared photon energies provide key parameters for waveguide second-harmonic generation. The latter is finally demonstrated by grating-coupling femtosecond pulses into the slabs. In the future, nano-optical imaging may be used to directly capture the frequency conversion process during the propagation of the modes. Our work demonstrates the feasibility of compact frequency converters and examines the tunings knobs that enable optimized coupling into layered waveguides.

O 84.3 Thu 11:30 H24

Imaging Infrared Materials Excitations with Sum-Frequency Spectro-Microscopy — •Niclas S. Mueller[1], Alexander Fellows[1], Richarda Niemann[1,2], Andrew E. Naclerio[2], Ryan A. Kowalski[2], Ben John[1], Katayoun Gharagozloo-Hubmann[3], Soeren Wasserroth[1], Guanyu Lu[2,4], Joshua D. Caldwell[2], Piran R. Kidambi[2], Martin Wolf[1], Martin Thaemer[1], and Alexander Paarmann[1] — [1]Fritz-Haber-Institute of the Max-Planck-Society, Berlin, Germany — [2]Vanderbilt University, Nashville, TN, USA — [3]Freie Universitaet Berlin, Berlin, Germany — [4]Northwestern University, Evanston, IL, USA

Nonlinear optical microscopy and spectroscopy are powerful tools to characterize interfaces and lower-dimensional materials. Here, I show two examples how we use infrared + visible sum-frequency generation (SFG) to image mid-infrared materials excitations with wide-field optical microscopy. The techniques provide combined spatial and spectral information, with sub-diffractional spatial resolution. 1. We visualize the propagation of infrared phonon polaritons in a metasurface of silicon carbide [1]. Through a combination of microscopy and spectroscopy, we observe the hybridization and strong coupling of propagating and localized polaritons, as well as the activation of edge states. 2. We visualize monolayers of hexagonal boron nitride, which are usually optically invisible. Resonant infrared excitation of phonons and heterodyne sum-frequency imaging enable us to image, both, the local topography and crystal orientation with unprecedented detail.

[1] Niemann, Mueller et al. Advanced Materials 36, 2312507 (2024)

O 84.4 Thu 12:00 H24

Towards Understanding Surface Restructuring: Automatic Process Exploration and Local Environment Analysis — •Patricia Poths, King Chun Lai, Francesco Cannizzaro, Christoph Scheurer, Sebastian Matera, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

The formation of surface oxides on the Pd(100) surface is a prototypical example of catalyst surface restructuring under *operando* conditions. Despite extensive investigation of this process, its mechanism and corresponding time scales remain unknown. By combining a Machine-Learned Interatomic Potential with our newly-developed Automatic Process Explorer (APE) [1], we study the O-induced restructuring of the Pd(410) vicinal surface as an example for step-edge initiation of oxide formation. We identify ∼3,000 unique elementary processes. Taking first steps towards a comprehensive mechanism, we analyze this rich library of processes utilizing a fuzzy classification methodology [2]. Many identified pathways involve non-intuitive complex collective motion of several atoms. These pathways can explain experimentally-observed restructuring, such as step bunching or microfaceting. Furthermore, the time scales of the majority of restructuring pathways are comparable to those of catalytic processes, implying significant coupling between surface restructuring and catalysis [3].

[1] Lai *et al.*, ChemRxiv, DOI:10.26434/chemrxiv-2024-jbzr7 .

[2] Lai *et al.*, J. Chem. Phys. **159**, 024129 (2023).

[3] Poths *et al.*, ChemRxiv, DOI:10.26434/chemrxiv-2024-36l28 .

O 84.5 Thu 12:30 H24

band engineering via metal substitution in porphyrin-graphene hybrid systems — •feifei xiang[1], yanwei gu[2], amogh kinikar[1], nicolò bassi[1], andres ortega-guerrero[1], carlo a. pignedoli[1], giovanni bottari[3], tomás torres[3], klaus müllen[2], pascal ruffieux[1], and roman fasel[1] — [1]Empa, Swiss Federal Laboratories for Materials Science and Technology, 8600 Dübendorf, Switzerland — [2]Synthetic Chemistry, Max Planck Institute for Polymer Research, 55128 Mainz, Germany — [3]Departamento de Química Orgánica Universidad Autónoma de Madrid Madrid 28049, Spain

Graphene and its derivatives serve as a versatile platform for finely tuning physical properties across a wide range. Expanding the potential applications of graphene to optoelectronics and gas sensing has driven the development of new materials including hybrid molecular systems that integrate porphyrins with graphene. On-surface synthesis provides a viable approach to achieving such well-defined hybrid structures. Here, we present the synthesis and characterization of porphyrin-graphene hybrid systems (chemrxiv-2024-z9rgh), in which the porphyrin units feature varying metal substitutions. The strong electronic coupling between porphyrin and tailored graphene nanostructures enables the metal center to play a significant role in modulating the band structure through charge doping, and to generate new hybrid states arising from d-π interactions, which allows long range spin coupling between magnetic metal centers. Such robust hybrid molecular systems hold promise in applications in p-n heterojunctions and paves the way towards achieving coherent control of spin chains.