

## O 88: Gerhard Ertl Young Investigator Award

Time: Thursday 10:30–13:00

Location: MA 005

**Invited Talk**

O 88.1 Thu 10:30 MA 005

**Syngas reactions on metal surfaces studied using scaling-relation-based kinetic Monte Carlo** — ●MIE ANDERSEN — Theoretical Chemistry, Technische Universität München, Germany

The complexity of catalytic reactions on surfaces calls for efficient means of estimating adsorption energies and reaction barriers, which are required input for a microkinetic model. One commonly used approach is to employ scaling relations [1], which are linear relations between the adsorption energies of atomic and molecular species on various (typically metal) surfaces, in combination with simple rate equations for the catalytic activity based on the mean-field approximation (MFA). In my talk, I will demonstrate the prospects of instead combining scaling relations with kinetic Monte Carlo simulations, which allow for predicting the catalytic activity while taking into account spatial correlations in the distribution of the adsorbates on the surface. For the specific case of methane synthesis from syngas on stepped metal surfaces, I demonstrate that MFA models may overestimate the catalytic activity of metal catalysts by several orders of magnitude due to the neglect of spatial correlations [2]. Finally, I will discuss recent and ongoing work aimed at (i) overcoming challenges related to a large disparity in the timescales of the surface processes, (ii) accounting for lateral interactions between the adsorbates, and (iii) dealing with more complex reaction networks such as the synthesis of higher alcohols on metal catalysts.

[1] F. Abild-Pedersen, *Catal. Today* **272**, 6 (2016)

[2] M. Andersen *et al.*, *J. Chem. Phys.* **147**, 152705 (2017)

**Invited Talk**

O 88.2 Thu 11:00 MA 005

**Catalytic reactivity of binary alloys studied by field emission techniques** — ●CÉDRIC BARROO, YANNICK DE DECKER, LUC JACOBS, and THIERRY VISART DE BOCARMÉ — Université Libre de Bruxelles

Field emission techniques are well-suited to study the dynamics of catalytic reactions occurring at the surface of a nanosized metal tip which represents a good model of a single catalytic nanoparticle. Here, field ion microscopy (FIM) is used to determine the structure of the alloys, and field emission microscopy (FEM) to study the dynamics of reactions during the ongoing processes. We studied the NO<sub>2</sub>+H<sub>2</sub> reaction on Pt-Rh. Previous experiments on Pt and Rh proved the presence and robustness of nonlinear dynamics, such as self-sustained periodic oscillations and propagation of chemical waves. On the Pt-Rh alloy, non-linear behaviors were observed: the characteristics of the oscillations lie between the features on pure Pt and Rh. The existence of periodic oscillations during the NO<sub>2</sub>+H<sub>2</sub> reaction on Pt, Rh and Pt-Rh samples suggest that the mechanism behind the occurrence of those oscillations is robust and seems to mostly depend on the reaction itself and less on the nature of the substrate. The mechanism of reaction is discussed. We also studied the N<sub>2</sub>O+H<sub>2</sub> reaction on Au-Ag, where N<sub>2</sub>O serves as a supplier of O(ads) since the ability to supply adsorbed oxygen is the key for activity/selectivity on gold surfaces. The reactive behavior can be observed via the appearance of new field emission patterns. These results prove the relevance of field emission techniques to study the catalytic activity of alloys.

**Invited Talk**

O 88.3 Thu 11:30 MA 005

**Imaging spin polarization and orbital character at surfaces: from the Rashba effect to topological Fermi arcs** — ●H. BENTMANN, H. MAASS, C.-H. MIN, and F. REINERT — Experimentelle Physik VII, Universität Würzburg

With the discovery of topological quantum states in solids, surface states have emerged from a niche phenomenon to a central concept in modern surface science and condensed matter physics, in general. Protected surface states with unconventional spin textures in momentum space are the hallmark of topological phases of matter being driven by spin-orbit interaction. Here, we shall present systematic spin- and

angle-resolved photoemission investigations of strongly spin-orbit coupled surface states [1-3], spanning the range from the Rashba effect over topological insulators to the exotic Fermi arcs in Weyl semimetals. We will show that these experiments -based on a careful consideration of the photoemission transition-matrix-element [1]- are able to yield a detailed insight into the momentum-dependent structure of the surface-state wave function, in terms of spin polarization [1], orbital character [2], and bulk penetration [3]. For the example of the Weyl semimetal TaP, we will discuss how the measured orbital character across the Fermi surface reflects the non-trivial topology of the Fermi-arc surface states.

[1] HB *et al.*, *Phys. Rev. Lett.* **119**, 106401 (2017).

[2] H. Maass, HB, *et al.*, *Nature Commun.* **7**, 11621 (2016).

[3] C. Seibel, HB, *et al.*, *Phys. Rev. Lett.* **114**, 066802 (2015).

**Invited Talk**

O 88.4 Thu 12:00 MA 005

**Tuning optoelectronic properties of silicon quantum dots via surface chemistry** — ●MITA DASOG<sup>1,2,3</sup>, JONATHAN G. C. VEINOT<sup>2</sup>, and NATHAN S. LEWIS<sup>3</sup> — <sup>1</sup>Department of Chemistry, Dalhousie University, Halifax, NS, Canada — <sup>2</sup>Department of Chemistry, University of Alberta, Edmonton, AB, Canada — <sup>3</sup>Division of Chemistry and Chemistry Engineering, California Institute of Technology, Pasadena, CA, United States

Over the last thirty years, quantum dots have become an indispensable tool for optoelectronic applications. The optical properties of colloidal quantum dots can be tuned through their size and shape. Among them silicon quantum dots (Si-QDs) have attracted attention due to their natural abundance and bio-compatibility. While few reports exist on size dependent emission from Si-QDs, the vast majority of them defy quantum confinement effect. In this talk, surface chemistry methodologies and luminescence in Si-QDs originating from surface states will be discussed. It was discovered that the emission can be tuned across the visible spectrum by changing the surface groups. The blue-to-green emission originating from oxynitride defect-states have shorter excited lifetimes and higher emission quantum yields compared to the bandgap emission in Si-QDs and the yellow-to-orange emission can originate from suboxide defect-states and have longer excited lifetimes and lower emission quantum yields. The Si-QDs can be further functionalized with surface molecules for bioimaging and explosives sensing.

**Invited Talk**

O 88.5 Thu 12:30 MA 005

**Carbon Dioxide Activation at Metal-Oxide Surfaces: A Compressed-Sensing Analysis** — ●ALIAKSEI MAZHEIKA<sup>1</sup>, YANGGANG WANG<sup>1</sup>, ROSENDO VALERO<sup>2</sup>, FRANCESC ILLAS<sup>2</sup>, RUNHAI OUYANG<sup>1</sup>, LUCA M. GHIRINGHELLI<sup>1</sup>, SERGEY V. LEVCHENKO<sup>1</sup>, and MATTHIAS SCHEFFLER<sup>1</sup> — <sup>1</sup>Fritz-Haber-Institut der MPG, Berlin 14195, Germany — <sup>2</sup>Universitat de Barcelona, Barcelona 08007, Spain

Conversion of carbon dioxide (CO<sub>2</sub>) to hydrocarbon fuels would allow for a sustainable energy utilization. CO<sub>2</sub> is a stable molecule, which requires an activation prior to conversion. It can be activated by adsorption on a solid surface. In this work, we employ compressed-sensing (SISSO) [1] and subgroup-discovery [2] approaches to find physically interpretable *ab initio* descriptors for energy and structure of CO<sub>2</sub> adsorbed at binary and ternary oxide surfaces. The descriptors include only properties of involved atomic species, bulk materials, and clean surfaces. We show that, contrary to the standard understanding, the O-C-O bending angle does not correlate well with the charge transferred to CO<sub>2</sub> for the whole data set. However, the subgroup discovery identifies a subset of surfaces for which this correlation is accurate. This subset is characterized by a more ionic character of the bonding between surface cations and O. Applying SISSO to this and the remaining subset independently, we obtain more accurate descriptors than for the whole data set.

[1] R. Ouyang *et al.*, arXiv:1710.03319

[2] B. Goldsmith *et al.*, *New J. Phys.* **19** 013031 (2017)