

O 57: Focus Session: Semiconductor Surface Chemistry – from Reaction Mechanisms to Well-Ordered Interfaces II

Time: Wednesday 15:00–17:00

Location: GER 38

Topical Talk O 57.1 Wed 15:00 GER 38
Incorporation of arsenic into silicon (001) and germanium (001) for atomic-scale device fabrication. — ●STEVEN R. SCHOFIELD — London Centre for Nanotechnology, University College London, 17-19 Gordon St, WC1H 0AH, United Kingdom

Atomic-scale electronic devices can be fabricated via the deterministic placement of individual donor atoms in semiconductors. The commonly-used technique involves the thermal decomposition of phosphine on the Si(001) surface and the patterning of a hydrogen resist layer for spatial control. However, it is now established that the desorption of phosphorus from the surface during the thermal incorporation anneal limits the scale-up of this method, e.g., for the fabrication of large numbers of qubits. In this talk, I will present exciting new work in our group demonstrating the excellent characteristics of arsine adsorption to both Si(001) and Ge(001) for the creation of atomic-scale devices. I will present combined scanning tunnelling microscopy and density functional theory work demonstrating that arsenic incorporates into the Ge(001) surface at room temperature when exposed to arsine, thus eliminating the need for the thermal anneal that is so problematic for phosphine on Si(001). Furthermore, I will present soft x-ray angle-resolved photoelectron spectroscopy (SX-ARPES) measurements of delta-doped layers in silicon that demonstrate higher confinement for arsenic layers compared to phosphorus layers. Our new results offer exciting opportunities for the fabrication of donor-based devices and their scale-up to the large numbers of qubits required for the fabrication of technological quantum devices.

O 57.2 Wed 15:30 GER 38
Controlling tip-induced reaction products of surface-adsorbed organic species on Si(001) — ●ALEXA ADAMKIEWICZ¹, TAMAM BOHAMUD^{1,2}, MARCEL REUTZEL¹, GERSON METTE¹, ULRICH HÖFER¹, and MICHAEL DÜRR^{1,2} — ¹Department of Physics, Philipps University Marburg, Germany — ²Institute of Applied Physics, Justus Liebig University Giessen, Germany

Tip-induced electronic excitation via STM can give access to alternative reaction channels beyond thermal activation. We showed for the cleavage of tetrahydrofuran (THF) on Si(001) that excitation by the tunneling electrons leads to new final products, which can be selectively addressed by the type of excitation [1]. Here, we present the influence of the detailed configuration of the organic adsorbates on tip-induced ether cleavage. Due to the linear molecular structure of diethyl ether (Et₂O), the adsorbates undergo fragmentation when cleaved, thus leading to a higher degree of freedom compared to the ring-shaped THF. The final products differ significantly for both species. Furthermore, we show that upon high voltage scans, covalently bound Et₂O fragments can be further reacted into more complex configurations which are thermally not accessible [2].

[1] Mette *et al.*, *Angew. Chem. Int. Edit.* **58**, 3417 (2019).

[2] Adamkiewicz *et al.*, *J. Phys.: Condens. Matter* **33**, 344004 (2021).

Topical Talk O 57.3 Wed 15:45 GER 38
Semiconductor surface chemistry towards hybrid interfaces with ab initio approaches — ●RALF TONNER-ZECH — Universität Leipzig, Leipzig, Germany

Exploiting concepts from molecular chemistry helps to improve our understanding of organic molecules reacting with semiconductor surfaces like silicon or germanium. Electronic structure analysis reveals concepts like Pauli repulsion or nucleophilic substitution reactions. Starting from this, hybrid interfaces can step by step be designed and analysed. The talk will focus on the associated challenges for theory encompassing model building, accuracy, adsorption kinetics and conceptual understanding. Finally, interface design from theory will be outlined.

[1] Pecher, Tonner, *WIREs Comput. Mol. Sci.* 2019, 9, e1401. [2] Luy, Molla, Pecher, Tonner, *J. Comput. Chem.* 2021, 42, 827. [3] Glaser, Meinecke, Freund, Langer, Luy, Tonner, Koert, Dürr, *Chem.–Eur. J.* 2021, 27, 8082.

O 57.4 Wed 16:15 GER 38
Oxygen vacancy occupancy influences oxygen evolution on

BiVO₄ — ●NICKLAS ÖSTERBACKA, HASSAN OUHBI, and JULIA WIKTOR — Chalmers University of Technology, Gothenburg, Sweden

Bismuth vanadate, BiVO₄, is one of the best semiconductor photoanode materials for photoelectrochemical water splitting. Oxygen vacancies are abundant in the material and greatly affect its photoelectrochemical properties. If present at the surface, these defects could additionally directly influence the oxygen evolution reaction. The electronic occupancy of the defect states may also play a role, but are seldom considered in mechanistic studies.

In this work, we use hybrid density functional theory to show that the surface oxygen vacancy in bismuth vanadate is most stable in its fully ionized state. We investigate how this affects the mechanics of the oxygen evolution reaction and compare the pathway with those involving the unionized surface vacancy and the defect-free surface. To quantify whether or not vacancy ionization is beneficial, the thermodynamic overpotentials required to drive the reaction are also computed and compared.

O 57.5 Wed 16:30 GER 38
Adsorption and photocatalytic inactivation of SARS-CoV-2 and virus like particles on the surface of anatase TiO₂(101)

— ●MONA KOHANTORABI¹, MICHAEL WAGSTAFFE¹, MARCUS CREUTZBURG¹, ESKO ERICK BECK¹, JOHANNES ROESSLER², REINHARD ZEIDLER², ALEXANDER HERRMANN³, MARTIN FEUERHERD³, GREGOR EBERT³, GABRIELA GUÉDEZ⁴, CHRISTIAN LÖW⁴, ROLAND THUENAUER⁴, THOMAS F. KELLER^{1,5}, BENEDIKT SOCHOR⁶, MATTHIAS SCHWARTZKOPF⁶, ANDREI CHUMAKOV⁶, STEPHAN V. ROTH⁶, ULRIKE PROTZER³, WOLFGANG HAMMERSCHMIDT², ANDREAS STIERLE^{1,5}, and HESHMAT NOEI¹ — ¹Centre for X-ray and Nano Science, DESY — ²Helmholtz Zentrum München — ³Technical University of Munich — ⁴Centre for Structural Systems Biology, DESY — ⁵Department of Physics, University of Hamburg — ⁶Deutsches Elektronen-Synchrotron

In this contribution, adsorption of SARS-CoV-2 and SARS-CoV-2 like particles (VLPs) were investigated on the surface of the model photocatalyst TiO₂(101). The samples were analyzed by grazing-incidence small-angle X-ray scattering (GISAXS), atomic force microscopy (AFM), transmission electron microscopy and X-ray photoelectron spectroscopy to obtain insight into the interaction of the SARS-CoV-2 and VLPs with the surface of TiO₂(101). Furthermore, the UV induced photo inactivation of adsorbed species was investigated. AFM and GISAXS results on the UV-treated sample suggested that adsorbed viral particles undergo damage and photocatalytic oxidation on the surface of TiO₂ which can affect the structural proteins.

O 57.6 Wed 16:45 GER 38
Ab initio description of surface restructuring and phase boundaries under realistic conditions — ●YUANYUAN ZHOU^{1,3}, CHUNYE ZHU^{1,2}, MATTHIAS SCHEFFLER¹, and LUCA M GHIRINGHELLI¹ — ¹The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft and IRIS-Adlershof of the Humboldt-Universität zu Berlin — ²School of Advanced Manufacturing, Guangdong University of Technology, Jieyang 515200, China — ³Department of Physics, Technical University of Denmark, Kongens Lyngby, 2800, Denmark

A reliable description of surfaces equilibria in a reactive gas is a prerequisite for understanding mechanism of heterogeneous catalysis. However, studying phase equilibria at *ab initio* level, is a formidable challenge, especially for systems with sluggish barriers. In this work [Zhou *et al.* *Phys. Rev. Lett.* 128, 246101(2022)], we introduce a fully *ab initio* approach to determine temperature-pressure ($T-p$) surface phase diagram and to evaluate phase equilibria of surfaces in a gas phase. For this purpose, our replica-exchange grand-canonical (REGC) method [Zhou *et al.* *Phys. Rev. B.* 100, 174106(2019)], is extended by evaluating the heat capacity, $C_v(T,p)$, as function of T and p , thus locating phase boundaries where $C_v(T,p)$ shows ridges. The approach is demonstrated by addressing open questions for the Si(100) surface in a H₂ gas phase. 25 distinct stable surface phases are identified, most of which have not been observed experimentally, so far. The results also show that Si-Si-bonds forming/breaking is the driving force behind the experimentally 3×1 and 2×1 phase transition.