MM 11: Topical session (Symposium MM): Hydrogen in Materials

Hydrogen interactions with materials surfaces (catalysts)

Time: Monday 15:45–17:00 Location: H 0107

Topical Talk MM 11.1 Mon 15:45 H 0107

Hydrogen transportation across palladium surfaces: Microscopic mechanism and control — •Markus Wilde, Satoshi Ohno, and Katsuyuki Fukutani — Institute of Industrial Science, The University of Tokyo, 153-8505 Tokyo, Japan

The ingress of H_2 into and the release of H_2 from the interior of H-absorbing metals, widely utilized in metal hydride storage, H_2 purification, and Pd-catalyzed C=C hydrogenation [1], still lacks atomic-level understanding. We here clarify the H_2 absorption mechanism at Pd single crystal surfaces through a unique combination of H depth profiling with ^{15}N nuclear reaction analysis [2] and thermal desorption spectroscopy. We resolve the long-standing paradox that although chemisorbed surface H is predominantly transferred into the Pd interior, the large potential energy difference between surface H and H in the Pd bulk does not materialize in the activation energy for H_2 absorption [3]. In contrast to Pd(100) and Pd(111), both defects and regular terrace sites of Pd(110) are active for H_2 absorption, implying that the H_2 absorption kinetics are sensitive to the surface 'openness'. Moreover, by manipulating the surface structure of Pd(110) through CO-induced (de-)reconstructions, we demonstrate the possibility to control the desorption dynamics of Pd-dissolved hydrogen in a wide range of temperatures (160-375 K) [4].

[1] M. Wilde, et al., Angew. Chem. Int. Ed. 47 , 9289 (2008) [2] M. Wilde, K. Fukutani, Surf. Sci. Rep. 69, 196 (2014) [3] S. Ohno, et al., J. Chem. Phys. 140, 134705 (2014) [4] S. Ohno, et al., J. Phys. Chem. C 119, 11732 (2015).

Topical Talk MM 11.2 Mon 16:15 H 0107

Hydrogen interaction with metal substrates studied from first principles — •Axel Gross — Institut für Theoretische Chemie, Universität Ulm, 89069 Ulm, Germany

The interaction of hydrogen with metals is of great technological importance in different areas such as hydrogen storage or H_2 production and heterogeneous catalysis. First-principles electronic structure calculations based on density functional theory (DFT) represent a reliable tool to elucidate atomistic details of the structures and processes resulting upon the hydrogen-metal interaction, but also to identify the underlying electronic factors determining the interaction strength. In this talk, I will illustrate this using several examples.

The subsurface penetration of hydrogen on precovered surfaces has been studied using ab initio molecular dynamics simulations [1] showing that concerted processes can significantly ease the absorption of hydrogen. It will furthermore be shown how the interaction of hydrogen with metal surfaces can be tuned by changing the structure and/or composition of bimetallic surfaces [2]. Finally, I will also briefly sketch the important role of metal-hydrogen interactions at electrochemical interfaces relevant for energy storage and conversion [3].

[1] S. Sakong, C. Mosch, A. Lozano, H.F. Busnengo, and A. Groß, ChemPhysChem **13**, 3467 (2012).
[2] S. Sakong, J.M. Fischer, D. Mahlberg, R.J. Behm and A Groß, Electrocatal. **13**, 530 (2017).
[3] A. Groß, F. Gossenberger, X. Lin, M. Naderian, S. Sakong, and T. Roman, J. Electrochem. Soc. **161**, E3015 (2014).

MM 11.3 Mon 16:45 H 0107

First-principles study of hydrogen related defects in titanium dioxide — •Mohsen Sotoudeh[1], Marian Bongers[2], Vladimir Roddatis[2], Jakub Čížek[3], Carsten Nowak[2], Martin Wenderoth[4], Peter Blöchl[1,2], and Astrid Pundt[2] — [1]Institute for Theoretical Physics, Clausthal University of Technology, Leibnizstr. 10, 38678 Clausthal-Zellerfeld, Germany — [2]Institut für Materialphysik, Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany — [3]Department of Low-temperature Physics, Charles University in Prague, V Holešovičkách 2, 18000 Praha 8, Czech Republic — [4]IV. Physikalischen Institut, Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany

Titanium dioxide (rutile) under hydrogen loading has been studied using first-principles calculations. In particular, we studied the defect electrochemistry near the interface with a metal. Defects related to oxygen vacancies and hydrogen have been investigated for different Fermi-levels. The band-gap underestimation in density-functional theory has been corrected. The nature of the electronic structure of the defects have been explored in detail. The defect concentration near the interfaces and their changes under hydrogen loading have been investigated. The calculated defect concentrations and the defect electronic structure explain experimental EELS spectra measured under hydrogen loading. We provide a mechanistic picture of the underlying chemical processes.

The work has been supported by the DFG through SFB 1073, projects B03, C03 and C06 as well via the Heisenberg grant PU131/9-2.