

A Theoretical Description of Photo-Catalytic Water Splitting on Metal-Decorated Oxide Surfaces

Harald Oberhofer, Daniel Berger, Markus Sinstein, Karsten Reuter

Chair for Theoretical Chemistry, Technical University Munich, Germany

e-mail: harald.oberhofer@tum.de

Efficient, sustainable production of molecular hydrogen---an promising alternative to batteries in terms of energy storage---is still an unsolved problem. Implementation of direct water splitting using only sunlight and suitable metal-oxide photo-catalysts so far has been hampered by poor photon absorption properties of the materials and low reaction efficiencies. To understand the microscopic processes involved in photo-catalytic hydrogen production we implemented an implicit solvent model and a solid state QM/MM embedding scheme based on ChemShell into the all electron DFT code FHI-aims.[1] This allows us to study defects and charged systems---as occurring in electron-hole driven water splitting---without any spurious interaction between periodic images, while at the same time yielding the correct electrostatic potential and solvent screening in the QM region.

In order to overcome the limitations of current water splitting setups we study the use of small metal clusters as co-catalysts, the microscopic effect of which is still poorly understood in literature. We develop an enhanced version of the thermodynamic approach pioneered by Nørskov and Rossmeisl,[2] of water oxidation reactions on metal clusters in the non-scalable size regime (less than 55 atoms) and compare with the bare extended oxide surface.[3]

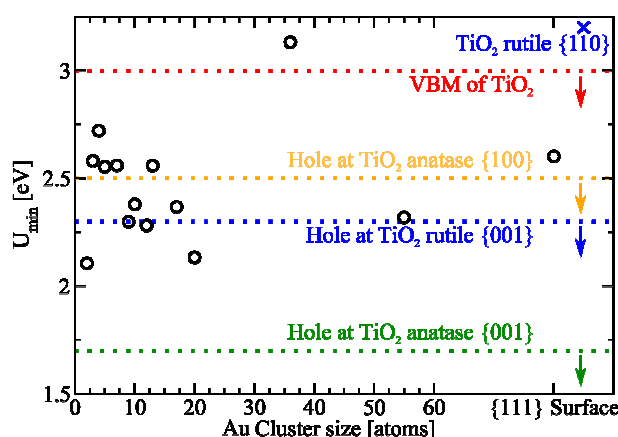


Fig 1: First screening results for TiO₂ nano-patterned with Au nano-clusters. Clusters lying below a certain dotted line are predicted to be catalytically active on that surface.

References

1. V. Blum *et al.*, Comp. Phys. Commun. **180**, 2175 (2009)
2. A. Valdes *et al.*, J. Phys. Chem. C **112**, 9872 (2008)
3. H. Oberhofer, K. Reuter, J. Chem. Phys. **139**, 44710 (2013)