Water Oxidation on WO$_3$(001) – An Embedded Cluster Study

Thomas Teusch[a] and Thorsten Klüner[a]

[a] *Carl-von-Ossietzky University Oldenburg, 26131 Oldenburg, Germany*
thomas.teusch@uni-oldenburg.de

Photocatalytic water splitting is a cutting-edge topic nowadays since there is no environmental friendly and efficient way known for hydrogen production. Tungsten trioxide (WO$_3$) is a promising material for photoanodes due to its many advantages like the narrower band gap compared to TiO$_2$, which allows for absorption of a large portion of the solar spectrum. Furthermore, the deep valence band position makes the water oxidation thermodynamically easier and the high electrical conductivity leads to good electron transport properties [1,2].

The aim of this contribution is to understand the involved mechanism of the water oxidation process on an atomistic level. To achieve this, we use hybrid DFT calculations with periodic boundary conditions [3] and multiconfigurational methods in terms of CASSCF [4]. The latter allows for the calculation of specific excited states in the relevant degrees of freedom.

We present results of the oxidation process by studying both ground and charge transfer states using an embedded cluster model. These results provide a molecular view into the water oxidation process and can also be used for high-precision wave-packet quantum dynamics which will allow for new time-resolved insights.

Since the wave packet dynamics require a very dense grid of potential points and the high-dimensional potential energy surface exhibits a too complex topology that can no longer be mapped using analytical expressions, we utilize machine learning as fitting procedure [5]. Applying state-of-the-art methods like dropout [6] and residual connections [7] we achieve a much more accurate and topology-preserving fit than using conventional neural networks.

References
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