Session of Focus Materialchemie – Wednesday, **5.6.2024** 16:00 – @ Seminarraum Lehar 01 (TU-Wien, Getreidemarkt 9, BC, OG. 01, room A46) – join us on ZOOM (ID: 983 0066 2349)

Layered perovskite oxides and perovskite oxynitrides: Interconversion, photocatalysis and stability

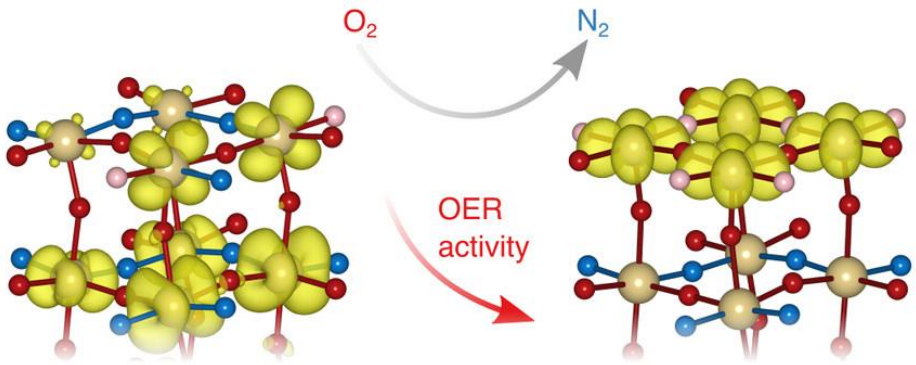
Ulrich Aschauer

ulrichjohannes.aschauer@plus.ac.at

Department of Chemistry and Physics of Materials, University of Salzburg

Due to their reduced band gap compared to oxides, perovskite oxynitrides are promising materials for visible light photocatalysis. Oxynitrides are commonly produced via ammonolysis from layered oxide materials. The ammonolysis process and the resulting physicochemical properties of the oxynitride are, however, poorly understood. Moreover, while the perovskite oxynitride band structure may be suitable to absorb visible light and drive the water-splitting half-reactions, their low catalytic activity and stability under application conditions remains a challenge.

Based on density functional theory, we develop a microscopic mechanistic understanding of the layered oxide to oxynitride conversion and investigate the role of crystal defects on the process and the resulting properties [1]. We will then explore the stability and reactivity of both the oxide [2] and the oxynitride [3] class of materials under photocatalytic application conditions. Our findings show that surfaces of both material classes will undergo structural changes in contact with an aqueous environment and under light irradiation, that severely impact their catalytic activity. Based on our computational results, we propose possible remedies to suppress the degradation and extend the lifetime of these photocatalysts.



Substitution of nitrogen by oxygen at a $SrTaO_2N$ surface occurs spontaneously under oxygen-evolution reaction (OER) conditions, negatively impacting the OER activity.

[1] Ricca C., Blandenier T., Werner V., Wang X., Pokrant S., Aschauer U., "*Conversion of $La_2Ti_2O_7$ to $LaTiO_2N$ via ammonolysis: a first-principles investigation*", **Phys. Chem. Chem. Phys.**, 25, 20575–20584 (2023).

[2] Bouri M., Niederhauser N., Künzli B., Amsler M., Aschauer, U., "*Oxygen Evolution Reaction Activity of $Sr_2Ta_2O_7$ and $Sr_2Nb_2O_7$ Surfaces*", **J. Phys. Chem. C**, 126, 6556–6563 (2022).

[3] Ouhbi H., Aschauer U. "*Nitrogen Loss and Oxygen Evolution Reaction Activity of Perovskite Oxynitrides*", **ACS Mat. Lett.** 1, 52–57 (2019).