Density functional theory study of anion ordering and chemical composition of different $LaTiO_2N$ surface orientations

S. Ninova[1], U. Aschauer[1]*

[1]University of Bern

Perovskite oxynitrides are a promising class of materials for photocatalytic water splitting under solar light.[1-3] They have the advantage of a reduced band gap with respect to the pure oxides, however at the cost of diminished stability. Perovskite oxynitrides are characterised by a partial order of oxygen and nitrogen atoms on the anion sublattice. In the bulk several theoretical studies find cis-positioning of the N-atoms to be energetically most favourable.[4-5] The anion ordering at the surface and its resulting chemical composition, however, are still elusive, despite its direct implication on the water-splitting mechanism on oxynitride surfaces.

We perform density-functional theory calculations on one member of the perovskite oxynitride class, $LaTiO_2N$, to deduce the O/N distribution for a variety of surface orientations and establish the chemical composition of the various surfaces and terminations. Based on these results it is possible to deduce trends for the reactivity of different surface orientations, which we correlate with experimental results on thin films with different orientations.

[1] Stefan G. Ebbinghaus, Hans-Peter Abicht, Richard Dronskowski, Thomas Müller, Armin Reller, Anke Weidenkaff, *Progress in Solid State Chemistry*, **2009**, 37, 173-205
[2] Tsuyoshi Takata, Chengsi Pan, Kayunari Domen, *Science and Technology of Advanced Materials*, **2015**, 16, 033506
[3] Asako Kasahara, Kota Nukumizu, Go Hitoki, Tsuyoshi Takata, Junko N. Kondo, Michikazu Hara, Hisayoshi Kobayashi, Kazunari Domen, *Journal of Physical Chemistry A*, **2002**, 106, 6750-6753
[4] Masamoto Yashima, Mio Saito, Hiromi Nakano, Tsuyoshi Takata, Kiyonori Ogisu, Kazunari Domen, *Chemical Communications*, **2010**, 46, 4704-4706`
[5] Spencer H. Porter, Zhenguo Huang, Patrick M. Woodward, *Crystal Growth and Design*, **2014**, 14, 117-125