Electronic Structure of La_xSr_{1-x}FeO_{3-δ} Probed by X-ray Absorption and Resonant Inelastic Soft X-ray Scattering

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For electrochemical water splitting, the oxygen evolution reaction (OER) is considered the efficiency-limiting factor. Therefore, the development of efficient catalytic materials for the OER has been a major research focus in recent years, with $La_xSr_{(1-x)}FeO_{(3-\delta)}$ (LSFO) being a promising candidate. In this material, with the perovskite (ABO₃) crystal structure, the oxidation state of Fe can be "tuned" by changing the La^{3+} to Sr^{2+} ratio. Increasing the Sr content (while keeping the oxygen content constant) increases the nominal amount of Fe⁴⁺ present in the material and increases the electrocatalytic activity.

We present a detailed study of the electronic structure of ozone-treated LSFO with different La/Sr ratios, by employing Fe L_{2,3} X-ray Absorption Spectroscopy (XAS), collected at the X-SPEC beamline at the KIT Light Source, and O K-edge Resonant Inelastic X-ray Scattering (RIXS) data, measured with our SALSA end station at Beamline 8.0.1 of the ALS. The spectra are analyzed with the help of Bethe-Salpeter Equation (BSE) calculations using the OCEAN code, painting a detailed picture of the electronic structure of LSFO as a function of the La/Sr ratio. In our contribution, we will show that the covalency between Fe and O increases with increasing Sr content and will discuss the applicability of the oxidation state concept for this materials class.



Figure 1. a) Coherent fraction of the experimental O K RIXS map and b) BSE calculated RIXS map of LaFeO₃. The BSE-calculated map captures all features of the experiment and gives insights in the electronic structure of the investigated material.