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Pulsed Laser Deposition: Preparing Model Systems for Studies on Energy Materials

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欢迎广大科研人员和研究生参与讨论!

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报告摘要

Fundamental understanding material properties and reactions of energy materials can often be very well studied by large facility techniques, e.g., at synchrotrons or neutron sources, as unique information can be obtained in this way. A number of these methods require the application of well-defined samples, controlling crystallinity, roughness to interface quality. These requirements can often be fulfilled by thin films. We apply pulsed laser deposition (PLD) to create these thin films to utilize complementary techniques, ranging from neutron reflectometry (NR) to grazing incidence X-ray absorption spectroscopy (GIXAS), and angle resolved photon emission spectroscopy (ARPES). One important aspect of our research is to increase our understanding of the pulsed laser deposition process and its possible shortcomings, such as deviations in composition. Here we apply a number of analytical techniques, e.g., mass and emission spectroscopy, which revealed the origin of the loss of elements in certain materials.

One of the material systems which we study are oxynitrides that are applied as photoanodes in photo-electrochemical water splitting. Shortcomings of this material class are a fast decay in activity over the first few electrochemical cycles and a decay on the long term. While the long-term decay is possibly related to a degradation of the material, i.e., a loss of nitrogen, the fast decay is not really understood, and therefore also no approach can be envisioned how to overcome this problem. We studied the fast decay of the material (and first approaches how to prevent this) by using thin films as model system. For this approach we developed a method on:

- How to deposit oxynitrides with well-defined oxygen content and crystallographic orientation by PLD using NH_3 as reactive gas component on conducting substrates.
- Design a cell for in-situ NR and in-situ/operando GIXAS.
- Measure the thin films before/after photoelectrochemical operation with NR and ARPES and before/after/during operation using GIXAS.

We could detect a surface modification, i.e., a change in density, by NR in the range of 3 nm, while XAS was utilized to analyze changes in oxidation state (order) for the different elements. A change of oxidation state of the A cation was detected, while the B cation (here for LaTiO_xN_y), which is normally assumed to be the active site, undergoes local disordering. This surface modification reduces the overall water splitting activity, but we could identify a co-catalyst, which suppresses these modifications. We could also identify critical steps in the water splitting mechanisms, where during surface modifications the formation of NO_x competes with the oxygen evolution. Using ARPES we could finally identify an electron accumulation layer at the surface as another mechanism for decreasing the activity.

Now we are working on approaches to mitigate the identified degradation mechanisms.

Without highly defined, high quality PLD films it would have not been possible to utilize the large facilities, and therefore to identify (mitigate) the origins of activity decay of these oxynitrides for water splitting.

报告人简介



Prof. Thomas Karl Michael Lippert is a professor in Department of Chemistry and Applied Biosciences, Laboratory of Inorganic Chemistry, ETH Zurich, Deputy Laboratory Head of the Laboratory for Multiscale Materials Experiments, and acting Laboratory Head since 7/2020, Group leader of Thin Films & Interfaces Group at the Paul Scherrer Institute, (formerly Materials) , PI principal investigator of the World Premier International Research Center-International Institute for Carbon-Neutral Energy Research in Kyushu University, president of the senate of the European-Materials Research Society (E-MRS) (1/2016-12/2017) and president of E-MRS (1/2014-12/2015).

His research are functional thin oxide films deposited by laser ablation, especially for renewable energy applications, such as fuel cells and batteries, but also with special properties (multiferroic), plasma spectroscopy (mass and emission) applied during laser ablation and thin film deposition, analysis and characterization of thin films using various spectroscopies (UV-Vis-NIR, Raman, Raman-microscopy, mass spectrometry and emission spectroscopy) including surface analysis by secondary ion mass spectrometry (SIMS) and application of laser ablation for fundamental studies related to structuring and laser transfer.