EUROPEAN SYNCHROTRON RADIATION FACILITY

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ESRF

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Combining Transmission Surface Diffraction and Laser Induced Fluorescence During Self-Sustained Reaction Oscillations	**Experiment number**: EV-255
Beamline: ID31	**Date of experiment**: from: 22.11.2017 to: 28.11.2017	**Date of report**: 28.02.2018
Shifts: 18	**Local contact(s)**: Jakub Drnec (and Tim Wiegmann)	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

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Report:

The original proposal intended the simultaneous study of model catalyst samples during operando CO oxidation reactions by combining the techniques of Transmission Surface Diffraction (TSD) and Planar Laser Induced Fluorescence (PLIF). In contrast to the original proposal, however, we did not combine TSD with PLIF, but we combined TSD with optical surface LED reflectance instead. The reason for this deviation from the proposed experiment was an unforeseen lack of experimentalists for the PLIF technique.
In optical surface LED reflectance a LED light is reflected from the sample surface and detected by a 2D camera. If the surface roughens or changes its optical properties, for example due to the formation of an oxide, the intensity of the reflected light detected by the camera changes. Here we wanted for the first time to correlate this change in reflected intensity to the surface structure of various model catalysts as determined by TSD.
The experiment was performed at the high energy beamline ID31 (E=80 keV) using the Pilatus 2M detector and our own operando catalysis chamber (comparable model to the one used at beamline ID03 but with a vertical sample heating stage allowing for TSD and a sapphire dome allowing for transmission of the LED light) with the optical surface LED reflectance set-up mounted on top of the chamber.
In the first part of the experiment, we intended to study MgO(001)-supported epitaxially grown Rh nanoparticles and epitaxially grown Rh thin films. Unfortunately, we could not detect any proper Bragg peak signals from these nanoparticles or thin films, which probably

can be traced back to problems we had during the sample preparation in our homelab in Lund, resulting in a low sample quality and which requires further inspection.

In the second part of the experiment we studied a Pd(001) single crystal which had a special cut-out drilled at its backside resulting in a 200 micrometer thin Pd area allowing for the transmission of the x-rays. For three different constant temperatures during reaction conditions for CO oxidation, we combined TSD and LED reflectance while running gas ramps from oxygen understoichiometry to oxygen overstoichiometry and back to oxygen understoichiometry. Moreover, we performed temperature ramps while keeping the CO/O_2 gas flow ratios during CO oxidation constant. Towards the very end of the experiment, as described in the proposal, we combined TSD and LED reflectance to study self-sustained reaction oscillations.

These measurements allowed for correlating changes in the LED reflectance intensity to the appearance/disappearance of epitaxial PdO bulk oxides as measured by TSD (see Fig. 1). This will help to identify the nature of the LED reflectance intensity changes since they can be correlated to structural changes on the model catalyst surface obtained by TSD.

Encountered problems:

The technique of TSD is based on looking at difference maps. This allows for following the appearance and disappearance of surface signals such as the ones that stem from oxides on the sample surface or from surface reconstructions. When substracting different images from each other we realized that the beam position sometimes varied between different images see central panel Fig. 1). A stable beam is, however, vital for the analysis of this kind of data. We ascribed the beam position instabilities to changes in the undulator gaps of neighbouring beamlines.

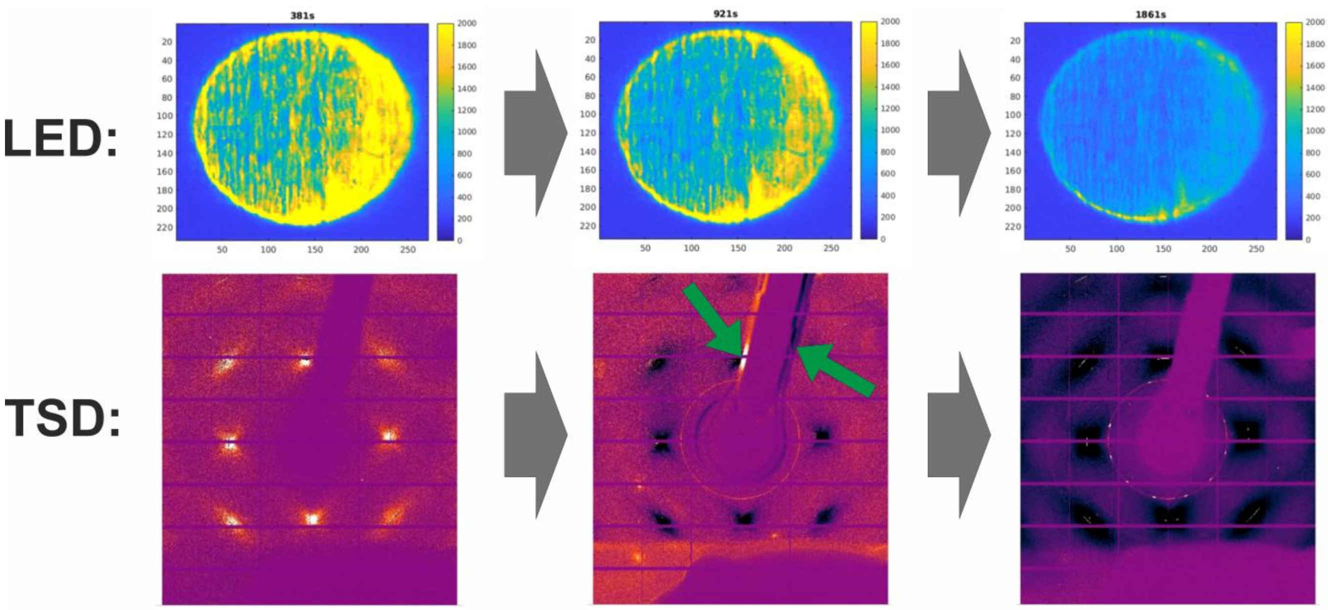

Fig. 1*: Correlation between optical surface LED reflectance (top panel) with TSD difference maps (bottom panel) in the transition from oxygen understoichiometry to high oxygen overstoichiometry (from left to right) on Pd(001) during CO oxidation. A decrease in the reflected LED intensity could be monitored during the formation of an epitaxial PdO bulk oxide. Please note the intensity variations in the central panel difference map around the beamstop shadow (green arrows) caused by beam position instabilities.*