EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

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

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal ("relevant report")

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *"preliminary report")*,
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as "relevant report(s)" in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

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.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number 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:** In situ study of the evolution of the defect landscape and the crystallinity of Cu_2O thin films upon laser annealing in different atmospheres.	**Experiment number**: A25-2-1079
Beamline: BM25	**Date of experiment**: from: 26 April 2023 at 08:00 to: 02 May 2023 at 08:00	**Date of report**: 14.07.2023
Shifts: 18	**Local contact(s)**: Dr. Juan Rubio Zuazo	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. Germán F. de la Fuente (*), Alejandro Frechilla Zabal (*) from INMA (CSIC-Universidad de Zaragoza)

Dr. D. Muñoz-Rojas (*), M. L. Johnston (*) and M. H. Okcu (*) from LMGP (CNRS-GRENOBLE INP)

Report:

This work originally aimed to perform in situ high energy X-ray diffraction and XAS analyses during the laser annealing of oxide thin films under different atmospheres. The data would be acquired from Cu_2O oxide thin films deposited on glass and Silicon by Spatial Atomic Layer Deposition (SALD) previously deposited at LMGP (Grenoble). The objective was to gain insight on the evolution in terms of crystallinity, phase and defects occurring during laser annealing under different atmospheres. XRD data would allow to follow the evolution during the laser annealing (amorphization due to partial melting followed by recrystallization vs. sintering and crystal growth, etc.), while local data obtained from the XAS results will provide insight on the effect of the laser treatment on the defect landscape in the films. The study aimed to complement XRD studies performed in the lab. The researchers involved in the experiment were, Dr. G. de la Fuente and M. A. Frechilla, from INMA (CSIC-UNIZAR) and Dr. D.Muñoz-Rojas, M. L. Johnston and M. H. Okcu from LMGP (CNRS-GRENOBLE INP):

Laser processing experiments were actually carried out in situ and operando, on Spatial Atomic Layer Deposition (SALD) prepared Cu_2O films, at the BM25 facility of ESRF between 26th and 30th April, 2023. In addition, ex situ experiments were also carried out on SALD deposited ZnO films during 1st and 2nd May, 2023, instead of the planned atmosphere controlled experiments, as lab results ready to be published were in accute need of the information that could be obtained from these experiments. For the former, a ns pulsed fiber laser was employed with beam (in situ) and line (operando) scan configurations. Figure 1 shows a picture of the in situ laser annealing set up that was used during the experiments. Finally, the beamtime was also used to acquire XAS data of NiO_x thin films deposited by SALD using a new precursor.

The purpose of the experiments was to obtain X-ray diffraction and EXAFS data unavailable with standard laboratory equipment, in order to study crystallographic arrangements and order, as well as other phenomena associated to the interaction of intense laser beams with solid films under very attractive configurations. These will enable scalability of processes and large area processing, as well as control of solid-state chemical transformations and microstructure. For obvious reasons, these methods are being protected from the IP point of view, and are subject to negotiations with an industrial partner, in relation to the SPRINT FET-OPEN project coordinated by the CNRS partner and participated by INMA. Figure 2 shows an example of low power and high power operando measurements, respectively.

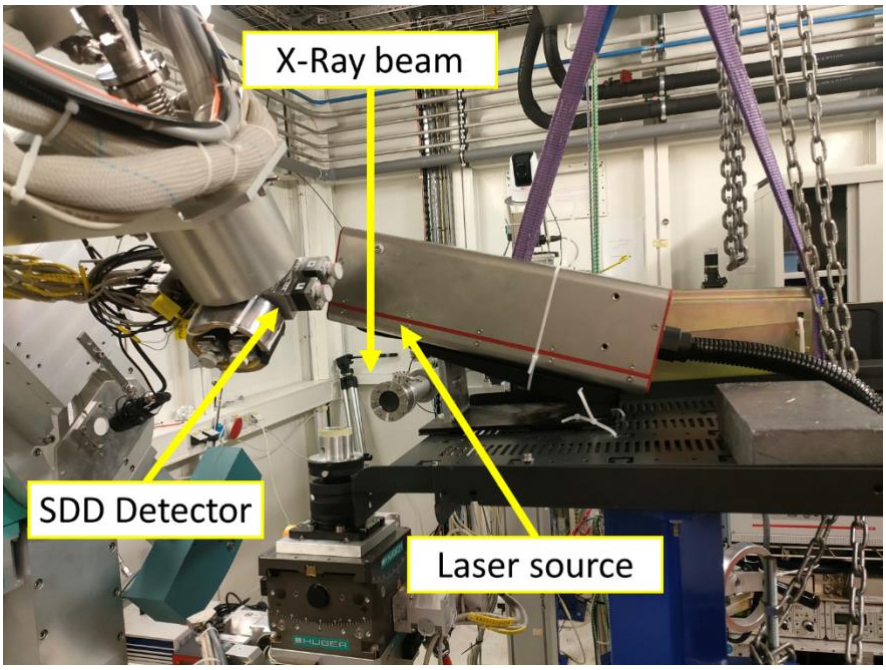

Figure 1: Set up developed for the in situ and operado laser annealing of SALD films in the beam line.

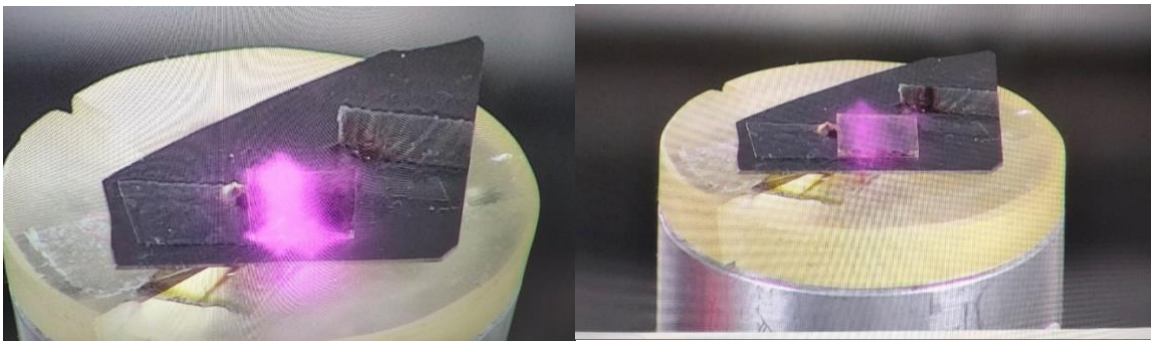

Figure 2. High power (left) and low power (rigth) operando measurement.

Figure 3 shows absortion spectra of 90 nm thick ZnO thin films after ex situ laser annealing of different pulse durations.

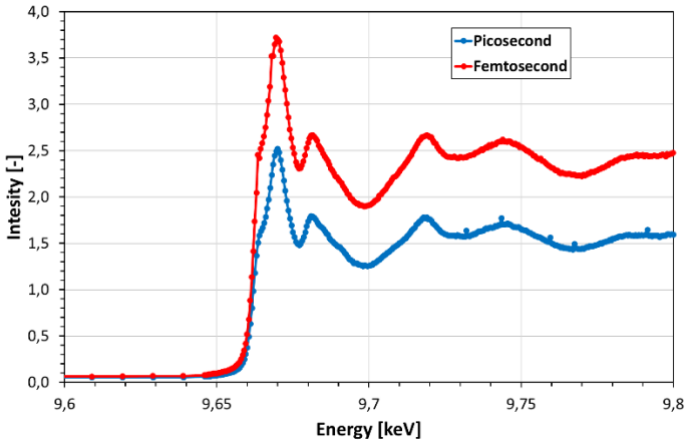

Figure 2. XAS data for ZnO thin films treated ex situ with different lasers.

The three young scientists (Ph.D. candidates) were heavily and enthusiastically involved in the experiments, along with two senior scientists (Professor level) and the Spline Staff (BM25), who were effectively involved in setting up the experiments and successfully solving any unexpected problem encountered during the installation and operation of the combined laser processing-irradiation set-up. Teamwork was excellent and yielded superb results which open the road to clarify and understand the phenomena taking place during laser surface treatment of SALD deposited films with respect to an increase in crystal order and the chemical changes induced by geometrically dosified laser irradiation.

The data obatined during the beam time is being treted and analysed and will be added to manuscripts that are in progress currently. Several manuscripts are expected as an outcome of these experiments, as well as new proposals regarding control of crystallinity and chemistry in laser surface treatment.