EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

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

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-operando hard X-ray nanoanalysis of quantum dot-based solar cells: The role of Sb in the capping layer	**Experiment number**: MA-5521
Beamline: ID16B	**Date of experiment**: from: 09/11/2022 to: 14/11/2022	**Date of report**:
Shifts: 15	**Local contact(s)**: Jaime Dolado	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Jaime Dolado [1], Malte Schwarz [2], Fernanda Malato [3], José Ulloa [2], Gema Martínez-Criado [1,3]

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Report:

Here, we report a correlative hard X-ray study of InAs/GaAsSb quantum dots (QDs) systems. Solar cells based on QDs are excellent candidates as novel photovoltaic devices due to their high efficiency (limit value of 63%, exceeding the 41% limiting value of ordinary solar cells).[1] An enhancement of 20% in power conversion efficiency has been demonstrated in these InAs/GaAsSb QD solar cells over the standard InAs/GaAs QD counterparts.[2] However, many questions are still open about the influence of Sb distribution in the band alignment and wavefunction topology. Using the hard X-ray nanobeam of ID16B station, InAs/GaAsSb QD solar cells with different Sb contents in the CL were studied regarding the Sb influence on the structure and the properties of the solar cell. The aim of the experiment was to obtain deeper insights into the origin of the efficiency enhancement with high statistics and superior correlations from large representative sample volumes.

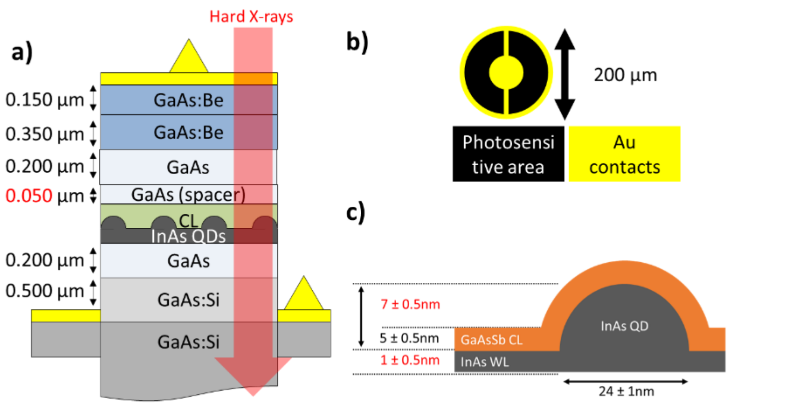

Figure 1: (a) Device Structure (b) Top View (c) QD-dimension schema.

For this experiment, we selected two InAs/GaAsSb QD solar cells (see Figure 1) with different Sb contents (10% and 28%). X-ray fluorescence (XRF) is applied to map the QDs areas, and X-ray absorption near edge structure (XANES) is measured to explore the local structure of In and Sb individually. The experiments were carried out using a beam lateral resolution of 80×60 nm^2. The XRF maps were taken at 30.5 keV (excitation energy above the Sb K-edge located at 30.49 keV). The spatially resolved XANES data were acquired in XRF detection mode around the In and Sb K-edge (27.94 keV and 30.49 keV, respectively).

Figure 2a shows the average XRF spectrum acquired on the sample with a 28% Sb content, using a pixel size of 50×50 nm^2 over a 900×900 nm^2 sample area and 1s/point integration time. Despite the elements present in the sample, the Mo peak is related to the monochromator used in the optical setup. The In and Sb XRF signal distributions are presented in Figures 2b and 2c respectively. The elemental distribution showed contrasting areas with different In intensity while the Sb distribution was very homogeneous. The regions with higher In signal indicate the accumulation of In, suggesting a higher amount of QDs. The assumption that high In content indicates a high amount of QDs is supported by simulations that consider the QDs distribution (measured by AFM, see Figure 2d) and the beam size (80×60 nm^2) to produce the image contrast expected in the experiment. The resulting simulation is presented in Figures 2e and 2f. As observed, the map resembles the experimental In XRF map (Figure 2b) corroborating that the measured contrast of In signal indeed enables localizing QDs areas.

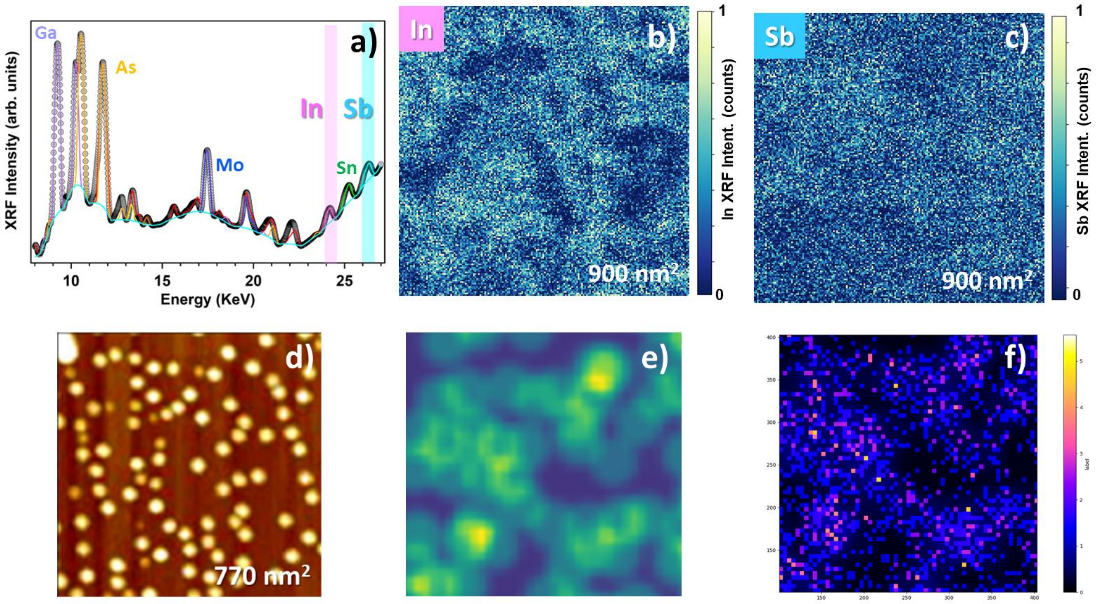

Figure 2: (a) Average XRF spectrum. XRF maps that depicts the XRF intensities of (b) In and (c) Sb in the device with a 28% Sb content. (d) AFM map of surface QDs. (e) and (f) Simulations of In distribution based on the AFM map shown in (a).

To study the effect of a possible Sb segregation in the capping layer, XANES around the K-edges of In (Figure 3a) and Sb (Figure 3b) was performed in the regions with high and low densities of QDs. The signal obtained for the In K-edge was weak and difficult to interpret beyond the location of the white line. Regarding the local environment of the Sb, no changes in coordination or electronic states are observed in the GaAsSb layers by the formation of QDs. A GaSb sample was measured around Sb K-edge as reference. Similar coordination can be observed between the GaSb reference and GaAsSb layers, with a slight shift towards higher energies in the case of GaAsSb, which may reflect a shorter scattering-absorption distance caused by the smaller bond lengths in the GaAs crystal. In addition, XANES spectra were also acquired in devices with different Sb content as displayed in Figure 3c, showing in both cases an intense narrow peak at around 30.50 keV attributed to the white line.

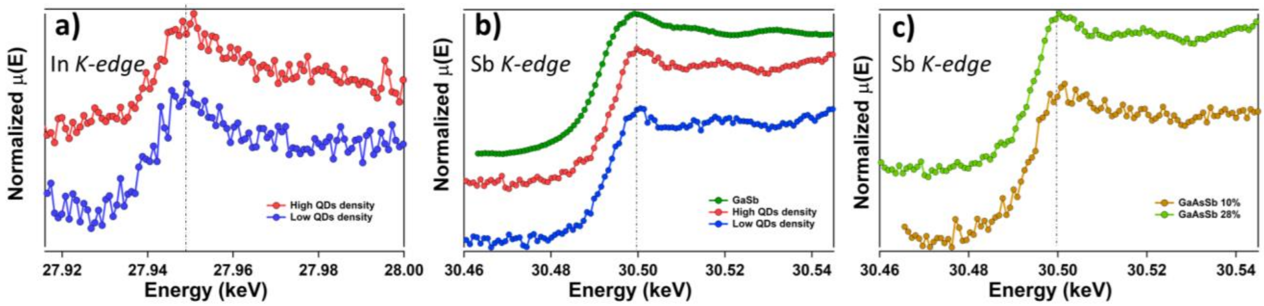

Figure 3: XANES data around the K-edge of (a) In and (b) Sb taken in regions with low and high QDs density in the device with a 28% Sb content. (c) XANES spectra around the Sb K-edge acquired in the devices with different Sb contents (10 and 28%).

The X-rays induced current (XBIC) was simultaneously measured with the XRF and XANES scans for further correlations. Due to technical problems related to the electronic configuration of the beamline, the XBIC measurements acquired during the experiment are not valid being necessary new experiments for the aimed *in-operando* analysis expected in the initial proposal. We anticipate repeating the XBIC measurements in the near future to complete the experiment.

References

1. X. Yang et al., Solar energy materials and solar cells, 113, 144-147 (2013).
2. AD. Utrilla et al., Solar Energy Mater Solar Cells, 159:282-289 (2017).