| ESRF | **Experiment title:** In-operando hard X-ray nanoanalysis on the impact of switching cycles on defects in III-Sb charge-storage devices for nonvolatile random-access memories | **Experiment number**: MA-5306 |
|---|---|---|
| **Beamline**: ID16B | **Date of experiment**: from: 28/06/2022 to: 04/07/2022 | **Date of report**: |
| **Shifts:** 15 | **Local contact(s)**: Jaime Segura | *Received at ESRF:* |


**Names and affiliations of applicants** (* indicates experimentalists):

Fernanda Malato*[1], Francisco Alvarado*[2], Xiuxin Xia*[3], Malte Schwarz*[4], Aurelia Trevisan*[5], Jaime Dolado*[6], Gema Martínez-Criado[1,6], Richard Beanland [2], Manus Hayne [3], Peter Hodgson[3]

*[1]Materials Science Institute of Madrid, [2] Warwick University, [3] Lancaster University, [4] Polytechnique University of Madrid, [5] Technology University of Eindhoven, [6] European Synchrotron Radiation Facility*


**Report:**

ULTRARAM is a non-volatile memory based on a triple barrier resonant tunnelling (TBRT) structure formed by multiple InAs/AlSb heterojunctions. With the TBRT structure, the device can be switched from a highly electrically resistive state to a highly conductive state by applying ± 2.5 V. Thus, non-volatility is achieved at extremely low switching energy per unit area.  Therefore, the paradox of universal memory, *i.e.* long storage time and ability to quickly write/erase data (add/remove charge) is solved by the tunnelling structure that provides a high-energy barrier when there is no bias applied but allows resonant tunnelling (*i.e.* transparent barriers) at program/erase (P/E) voltages of about 2.5 V. This is 10 times lower than flash, corresponding to a switching energy per unit area that is lower than DRAM and flash by 100 and 1000, respectively. The sample was grown by molecular beam epitaxy (MBE). First, an AlSb nucleation layer was deposited, in order to seed the growth of the GaSb buffer layer. Afterward, the III-V semiconductors epilayers were grown according to **Figure 1**. More information on the MBE growth of the sample is given in Ref. [1].

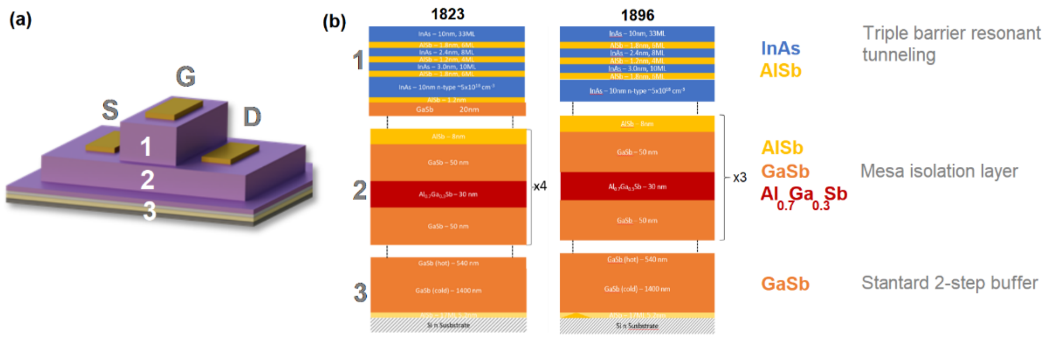


**Figure 1** – (a) Schematic of the ULTRARAM devices and the (b) layers configuration for fresh and failed devices (1823) and for the cycled device (1896).

This experiment aims to study whether any nanoparticles or clusters that deviate from the III-V stoichiometry are responsible for the detrimental performance. In addition, the impact of the number of P/E cycles on the local crystalline configuration is addressed by nano-X-ray absorption spectroscopy at Sb and In K-edges. The formation of defects under operating conditions or after/before multiple cycles is critical in transport device degradation. Three devices in different conditions (fresh, cycled, and failed) were studied at the nanoanalysis beamline of ID16B at the ESRF. The experiments were carried out using a beam lateral resolution of 62 x 67 nm$^2$ and excitation energies above the K edges of Sb (30.49 keV) and In (27.94 keV). The XANES spectra were acquired in XRF mode between 30.44 to 30.52 keV for Sb K-edge and between 27.85 to 28.05 keV for the In K-edge. The first results are present in the figures attached. To facilitate the application of electrical bias to switch the cells and thus allow for *operando* measurements, the devices were electrically connected to gold contact and wire bonded to a carrier chip, which could be electrically contacted from the sample holder.

**Figure 2** presents the explorative XANES data collected around the Sb K-edge for the devices in fresh, cycled, and failed states collected in the Gate. For comparison, the XANES of an Sb foil was measured as a reference at the Sb K-edge. The first results reveal that the device presents a uniform behaviour for the three devices studied, indicating no degradation around the Sb and In sites with cycling. Nevertheless, further simulations and references will be investigated to support the data analysis of results obtained in the experiments performed until now in order to understand the structural order and Sb coordination. Finally, **Figure 3** presents the XBIC signal which exhibits a dependence on time. As observed, an immediate increase in the current is observed by the X-ray beam excitation stabilizing to a maximum value. However, when the beam is off the current relaxes slowly reaching an intermediary plateau. Seconds before the XBIC starts to decrease until relaxes completely. This phenomenon was not yet been observed for this device and will be investigated for a deeper comprehension of the carrier transport in the ULTRARAM memories.

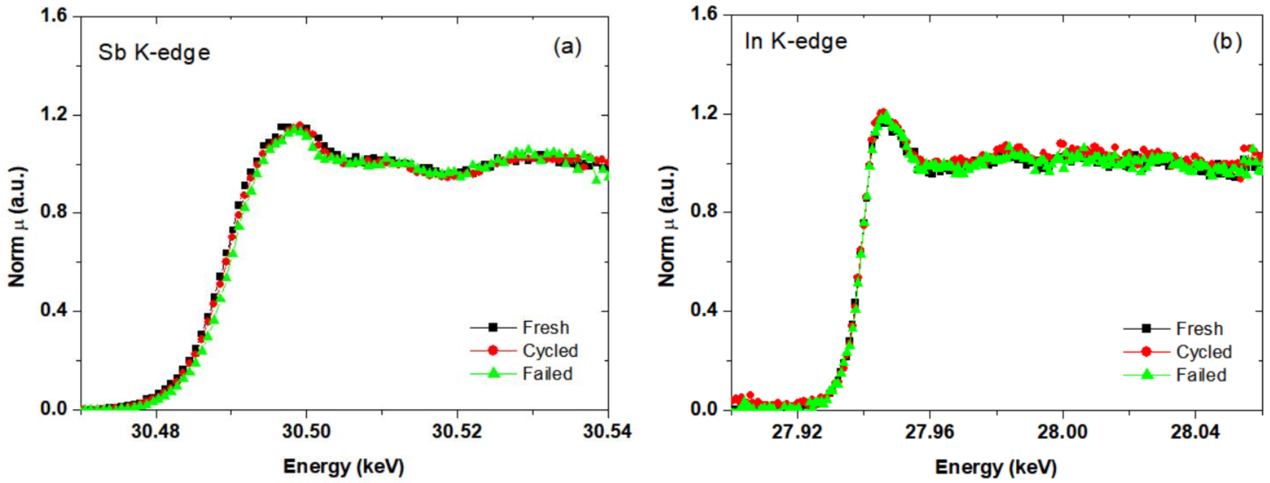


**Figure 2** – (a) Sb and (b) In K-edge XANES acquired from different devices (fresh, partially cycled, and cycled to failure) in fluorescence yield.

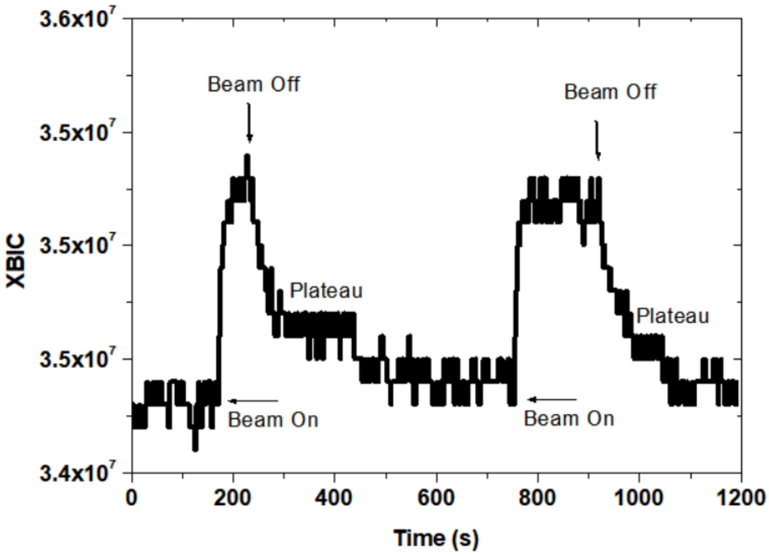


**Figure 3** – XBIC vs. Time dependence. The graph highlights the moments where the beam is exciting the device.

[1] *P. D. Hodgson et al. Advanced Electronic Materials,8:4,2101103, 2021*