ESRF	**Experiment title:** Filament Detection in Resistively Switching $SrTiO_3$ Thin Films by GISAXS	**Experiment number**: MA-1846
Beamline: ID 13	**Date of experiment**: from: 07.02.2014 to: 10.02. 2014	**Date of report**: 05.03.2014
Shifts: 9	**Local contact(s)**: Dr. Manfred Burghammer	*Received at ESRF:*

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Report: (Preliminary)

Owing to the short time span since the end of the beamtime, this report is only preliminary, accompanying a continuation proposal.

The experiment was carried out at ID13 with the aim of measuring the grazing incidence small angle X-ray scattering (GISAXS) signature of conductive filaments in resistively switching oxides. The filaments result from voltage-induced changes in the oxygen stoichiometry, with typical diameters on the order of 10 - 100 nm. The contrast between the filaments (typical stoichiometry $SrTiO_{3-\delta}$) and their surrounding matrix ($SrTiO_3$) is small (δ ca. 0.1) but sufficient to generate a small-angle scattering signal.

Such GISAXS experiments were performed for the first time at a nanofocus endstation, using an energy of 14.9 keV and a sample-to-detector distance of 1.53 m. To resolve the filamentary structures, a beam of 150 x 150 nm^2 cross section was used in scanning mode. The nanofocused beam was necessary to address single metal-insulator-metal structures (capacitor pads). The studied samples were thin $SrTiO_3$ films with various micron-sized metal electrodes produced by lithography. All measurements were performed ex-situ, e.g. the relevant electrical states (pristine/on/off) were prepared prior to the beamtime using external switching.

Sample Designation	Substrate	Active Layer	Electrode
Sub 03	10 x 10 mm^2 Nb:STO	20 nm Fe(0.5%):STO	10 nm Al
Sub 07	10 x 10 mm^2 Nb:STO	20 nm Fe(0.5%):STO	10 nm Al
CB 019	10 x 10 mm^2 Nb:STO	Substrate/electrode interface	5 nm Rh
HAPE 5	10 x 10 mm^2 Nb:STO	20 m Fe(5%):STO	5 nm Rh

Electrical Switching Results

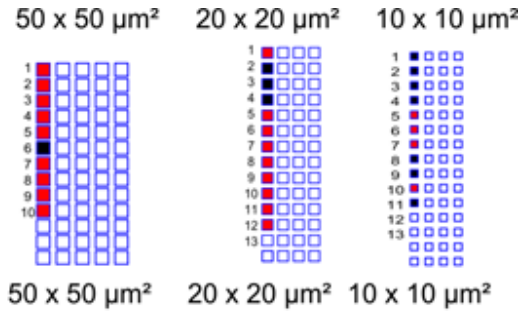

Fig. 1:
Investigated electrode arrays of sample Sub 03 (detail). The switching behaviour is color-coded (white = pristine, red = low resistive state, dark = switching not well defined).

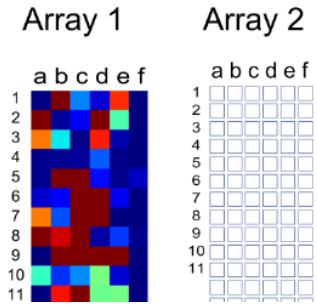

Fig. 2:
Electrode arrays of sample Sub 07 (detail) with their resistance color-coded (white = pristine, red = low resistance → blue = high resistance). The sizes of the capacitor pads are 20 x 20 and 50 x 50 µm^2.

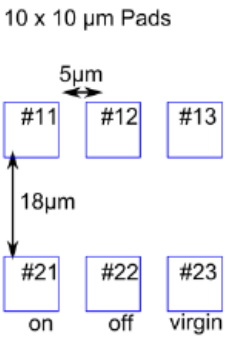

Fig. 3:
Investigated electrode arrays of sample CB 019 with their switching state indicated (detail from a large array; on state = low resistive state etc.).

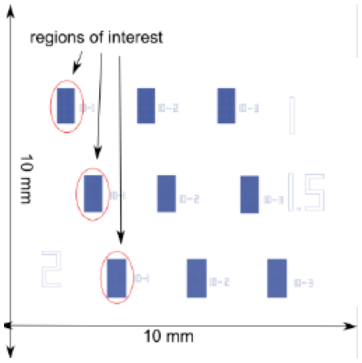

Fig. 4:
Investigated electrode arrays of sample HAPE 5. The areas consist of 10 µm x 10 µm capacitor pads. Top array: off state / middle: pristine / bottom: on state.

The samples investigated during the experiment are listed at the end of the previous page. They differ in electronic structure by Fe doping, but also with respect to the electrodes used. The relevant electrode arrays are represented in Fig. 1 – 4. The switching state of individual capacitors, which was determined prior to the ESRF experiment, is indicated in the figures.

GISAXS Results

The experiment was carried out at an incidence angle of 0.6°, resulting in a large footprint of the nanobeam of roughly 15 µm. This allowed for the systematic scanning of areas that match the electrode sizes very well. For each scanning point, a GISAXS pattern was recorded. Representative patterns for all measured samples are shown in Fig. 7 – 10. However, sample Sub07 is missing, since no scattering contrast could be observed between the electrodes and the substrate. The reason for this behavior is currently unknown. The patterns are arranged in Fig. 7 – 10 to represent the scanned area; hence the shape of the electrode arrays can be recognized. It should be noted that not the entire GISAXS pattern is represented in the figures, but only a characteristic (large) part of the pattern, which showed the largest variations during scanning.

Although it would seem at first sight that a correlation between local GISAXS data and electrical properties should be straightforward, the data evaluation currently going on has shown that this is not the case. The reason is that the nanobeam is highly coherent, which leads to a significant amount of speckle in the GISAXS data. This makes the otherwise straightforward comparison of different spots difficult, since many real structure effects have an impact on speckle patterns. On the other hand, the uniqueness of speckle patterns may also open up new possibilities to identify the filamentary channels. This avenue is currently explored.

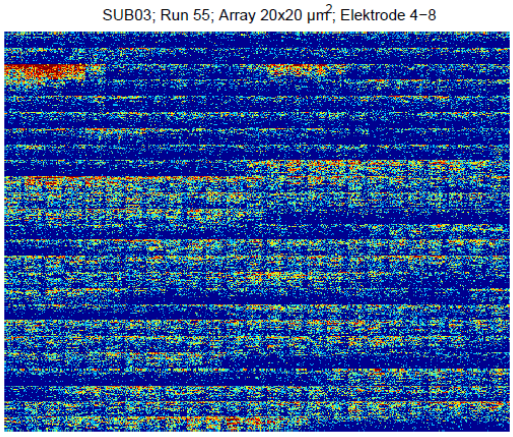

Fig. 5:
GISAXS scans of sample Sub 03.

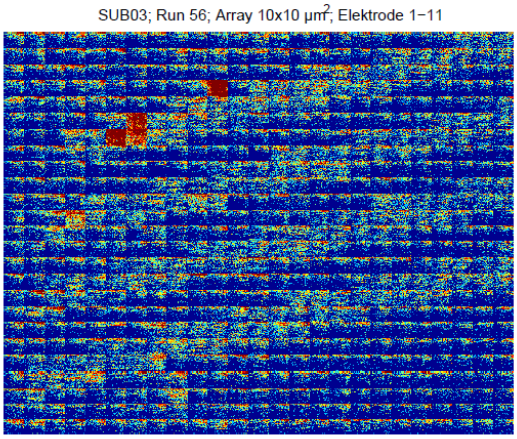

Fig. 6: GISAXS scans of sample Sub 03 (different set of electrodes).

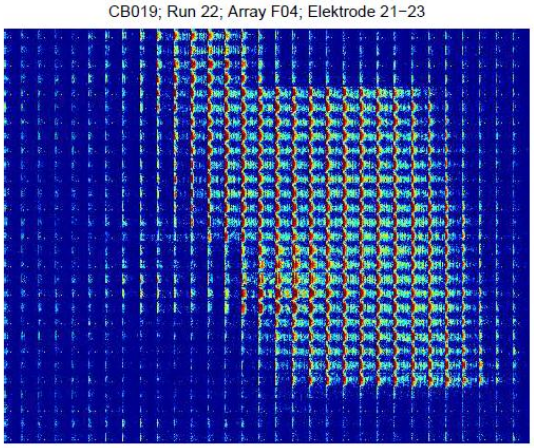

Fig. 7:
GISAXS scans of sample CB 019.

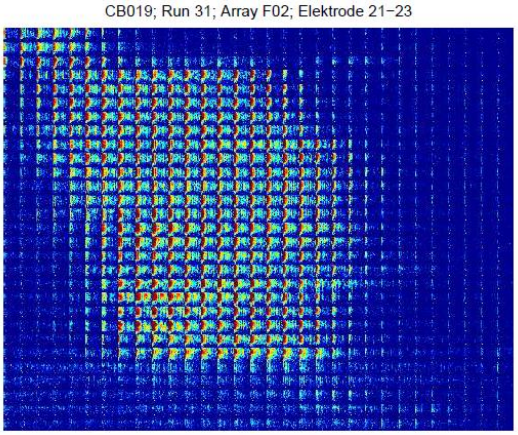

Fig. 8: GISAXS scans of sample CB 019 (different set of electrodes).

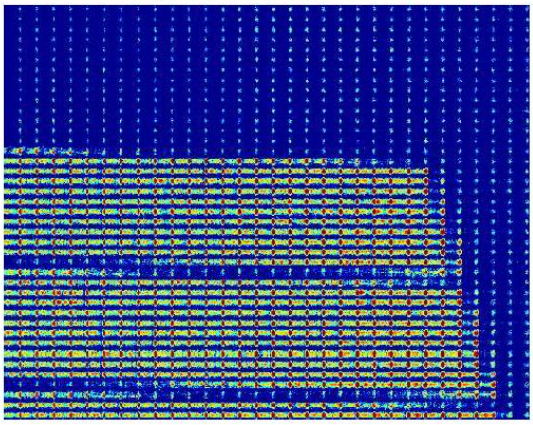

Fig. 9:
GISAXS scans of sample HAPE 5.

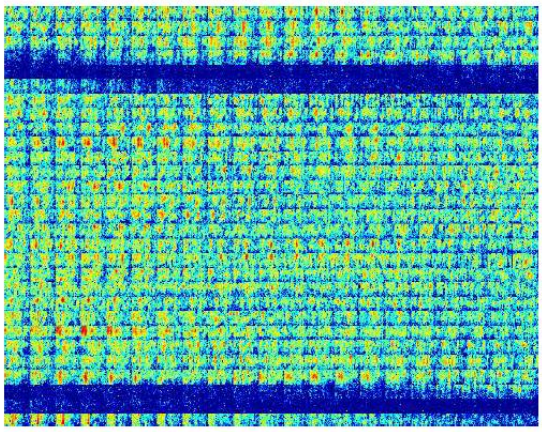

Fig. 10: GISAXS scans of sample HAPE 5 (different set of electrodes).