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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 using the **Electronic Report Submission Application:**

http://193.49.43.2:8080/smis/servlet/UserUtils?start

Reports supporting requests for additional beam time

Reports can now 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:** Structural and Dielectric Properties of Barium Strontium Titanate Thin Films	**Experiment number**: MA 288
Beamline: ID15	**Date of experiment**: from: 16th May 2007 to: 20th May 2007	**Date of report**:
Shifts: 9	**Local contact(s)**: Veijo Honkimaki	*Received at ESRF:*

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Report:

Experimental Setup

The sample analysed was a $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) single crystal thin film of 400 nm thickness grown on a single crystal MgO substrate of dimensions 10×10×0.5 mm. The sample was mounted in a 17x17 mm polymer carrier and wire bonded to electrical contacts (Figure 1(a)). The sample was loaded into a liquid nitrogen cooled cryostat constructed especially for this experiment by ESRF, containing high voltage lines for application of up to 1 kV to the sample (Figure 1(b)).

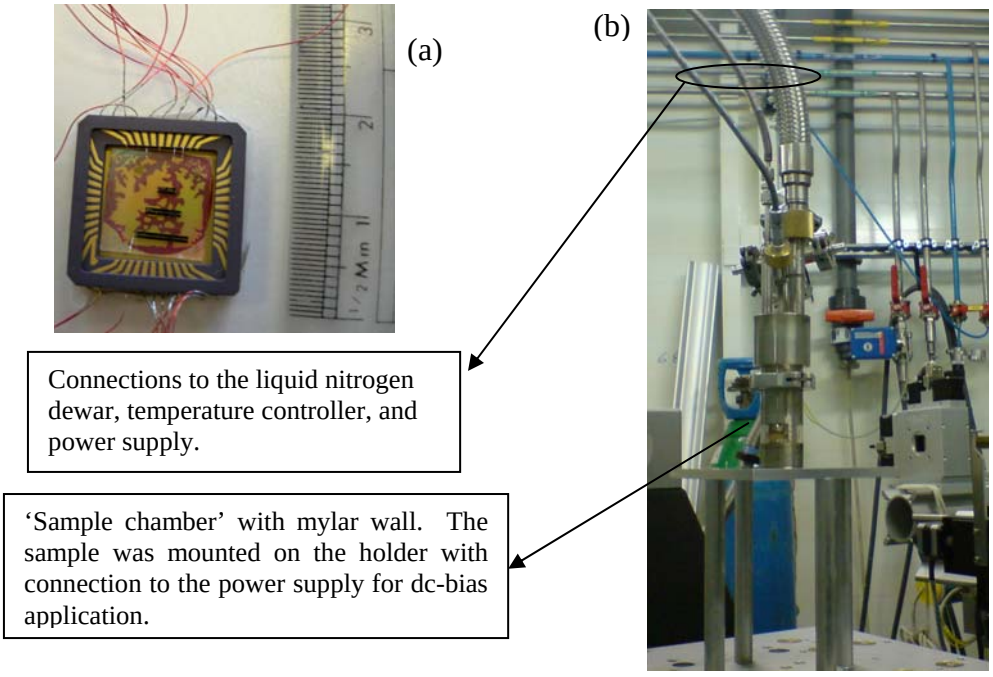

Figure 1 *(a) BST thin film on MgO substrate mounted on the sample carrier with wire bonding between the electrodes on the sample and the carrier, (b) The cryostat in place on the diffractometer cradle.*

The objectives of the experiment were to follow the evolution of the lattice parameters and strain in the film as a function of temperature through the tetragonal to cubic (ferroelectric to paraelectric) phase transition at ~240 K and to search for sensitivity of the structure to an applied electric field. The sensitivity to electric field was desired in particular to elucidate the mechanism of the field-dependence of the dielectric properties, which is our main research focus. To this end the high energy X-ray beam (70 keV) at ID15 was focussed to a microbeam of dimensions approximately 15 x 8 μm and directed at grazing incidence onto the sample between the silver electrodes on the top surface (Figure 2a). This positioning between the electrodes was achieved by monitoring the X-ray fluoresence from silver while scanning the sample beneath the beam, as shown in Figure 2(b).

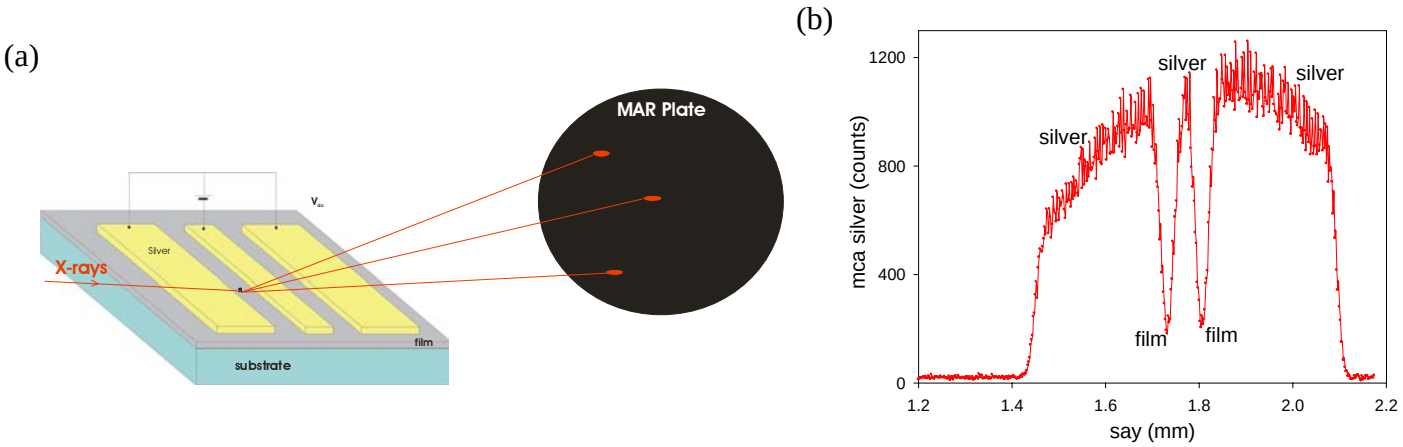

Figure 2 *(a) Grazing incidence of x-ray diffraction beam on the sample surface and diffracted onto the MAR plate, (b) Intensity of silver counts from x-ray fluoresecence across the sample surface*

Unfortunately, secondary scattering from the carrier required for connection of the electic bias lines in the cryostat to the sample was stronger than anticipated due to the need for grazing incidence geometry, so the sample had to be removed from the carrier. A new carrier without sidewalls has been designed in anticipation of a further beamtime allocation. We were, however, able to make a series of temperature scans in the absence of applied voltage.

Results

From the scanning profile in Figure 2(b), we could confidently locate the x-ray beam between the silver electrodes. Figure 3(a) shows an example of an x-ray diffraction pattern taken with the sample at 140 K. The presence of spots rather than rings proves that the BST film was single crystalline and had grown epitaxially on the MgO substrate. The pattern showed the (h00) and (h01) peaks of the BST film and (h00) peaks of the MgO substrate. The (200) BST and MgO peaks were almost coincident, due to the close matching of the lattice parameters of the two materials.

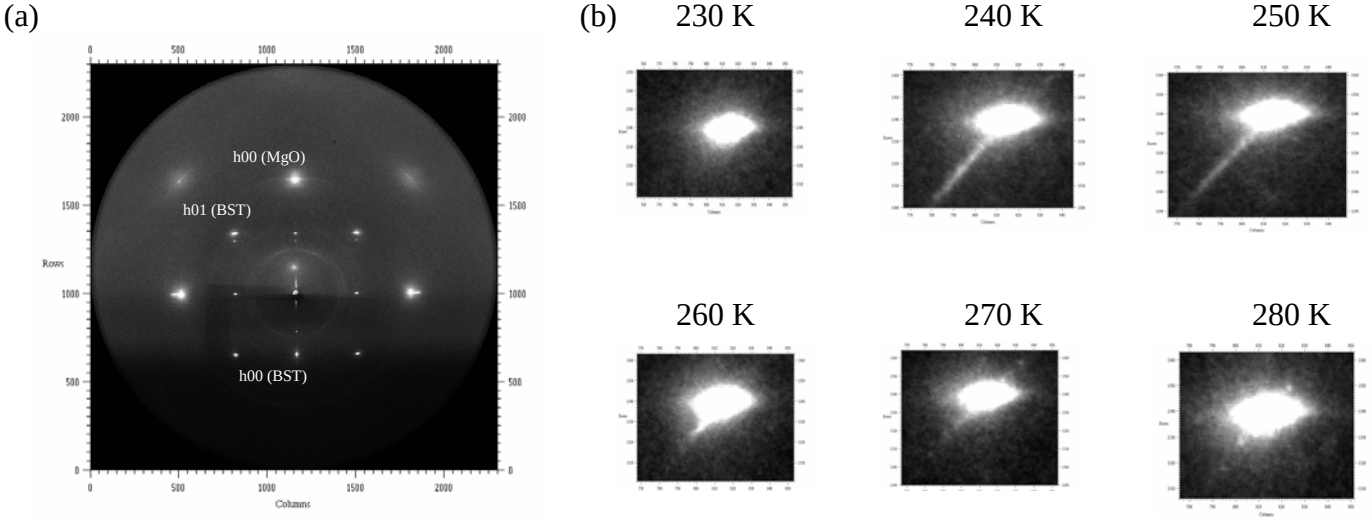

Figure 3 *(a) x-ray diffraction image of the sample at 140 K, (b) x-ray diffraction image of (h01) peak at 230 K – 280 K, showing the appearance of 'streaks' at 240 K – 280 K, demonstrating a phase transition of ferroelectric to paraelectric.*

The sample to detector distance was calibrated using an iron powder sample in place of the cryostat. To analyse the MAR-plate data, the radial location of the peaks with respect to the position of the straight-through beam was determined using the FIT2D software and these were converted to d-spacing values.

The phase transition from ferroelectric (tetragonal) to paraelectric (cubic) was observed on both heating and cooling. The onset of the transition was observed at 240 K, where the 'streaks' were observed on the (h01) peaks (Figure 3(b)). The 'streaks' started to disappear at 280 K. This phase transition at temperature range of 240 K – 280 K was consistent with that observed from our previous microwave measurements of the dielectric properties and from Raman spectroscopy.

The phase transition was also directly evident in the temperature dependence of the d-spacings of the (100) and (101) planes (Figure 4(a)). The lattice parameters were determined assuming a tetragonal structure below 240 K and cubic above. Figure 4(b) shows the temperature dependence of the a-axis and c-axis lattice parameters and the matching of the data at room temperature to previously published values. The corresponding data for the MgO substrate showed a linear thermal contraction, as expected.

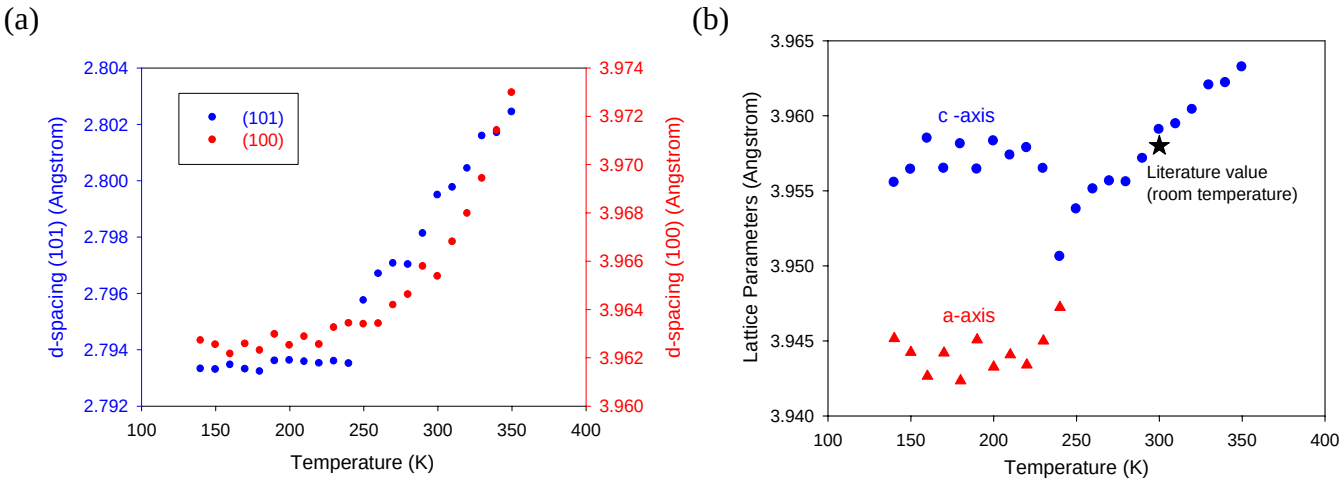

Figure 4 *Temperature dependence of (a) d-spacings of (h00) and (h01) peaks of BST film (b) temperature dependence of a-axis and c-axis lattice paramters.*

Discussion

The temperature dependence of the lattice parameters of the BST film has been determined from this experiment. The transition temperature and breadth of the transition were comparable with those deduced indirectly from dieletric measurements and Raman spectroscopy. However, the accuracy of the lattice parameters obtained from this preliminary experiment was not as high as desired, due in part to errors originating from our inexperience (this was our first detailed study of a single crystal thin film using synchrotron radiation). The errors and the steps that will be taken to overcome them in a future beamtime were:

- there were many overexposed peaks which created difficulty in finding the center and intensity of the diffraction peaks. This will be avoided by the use of slits and/or shorter exposure times to reduce the overall beam intensity;
- the tilt on the diffraction patterns, that caused difficulty in determining the width of the peaks directly from the FIT2D software. These tilts were temperature sensitive and so could not be corrected from the original calibration at room temperature. These could be removed by calibration from the substrate peaks if a wider range of incident and azimuthal angles are made available to probe more diffraction peaks;
- the resolution of the image pixel size on the setting of the MAR plate detector was barely sufficient to follow the changes in lattice parameter so the pixel size will be reduced from 150 μm to 100 μm (150 was thought sufficient and chosen to minimise file storage space). Further gains could be achieved by changing the data collection geometry, e.g. via increasing the sample to detector distance and/or moving the detector sideways to sample further out in reciprocal space.

With these changes, it should be possible to reliably track changes in the lattice parameters and strain under changing temperature and under electric field bias.