ESRF	**Experiment title:** Investigation of the effect of optical lithography on epitaxial layers, using Bragg tomography technique	**Experiment number**: HS-3636
Beamline: BM-05	**Date of experiment**: from: 28/01/2009 to: 03/02/2009	**Date of report**:
Shifts: 18	**Local contact(s)**: Dr. Eric ZIEGLER	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. Cristian MOCUTA, Synchrotron SOLEIL (*)

Dr. Antoine BARBIER, CEA-Saclay (*)

Dr Jean-Baptiste MOUSSY, CEA-Saclay (*)

Dr. Stefan STANESCU, Synchrotron SOLEIL (*)

Ms. Sylvia MATZEN, CEA-Saclay (*)

Dr. Julian STANGL, Linz University

Dr. Frédéric PETROFF, Unité Mixte de Physique CNRS/Thales, Palaiseau

Dr. Richard MATTANA, Unité Mixte de Physique CNRS/Thales, Palaiseau

Report: (preliminary)

Epitaxial metal/oxide magnetic tunnel junctions (MTJ) layered samples were used. Within a layered MTJ plain sample different junction sizes (10 – 150 μm) and shapes (squares, rectangles and disks) were lithographed; alignment marks (visible with a regular optical camera pointed at the sample) were used to put the beam on the area of interest. In a previous measurement we have shown that close to the junction edges the crystalline planes of the layers are tilted, with amplitudes ~1-3° [1].

In the first part of the experiment, a FReLoN 2D detector was used. The idea behind the experiment is the following: the sample is illuminated by a large parallel beam and the 2D detector, in θ-2θ specular geometry, is used to image (with high spatial resolution) the shape of the resulting Bragg peak (which will contain in this case the shape of the illuminated junction(s)). By taking images in Bragg conditions while rocking the sample incident angle (θ) it is expected to observe modifications of the distribution of the diffracted intensity on the area detector. It is then possible, in principle, to access the crystalline information (quality, tilts of planes, …) at each point of the sample, with a lateral resolution related to the one of the detector. If rocking scans around of the tilting angle perpendicular to the beam are performed, it is possible to access tilting planes information also for the junction edges parallel to the beam. Combining these 2 measurements, information from the whole junction area is accessed.

The individual layers in the junctions were chosen such that they have different lattice parameters and thus well separated Bragg peaks, and since the tilting effects are expected to be large (several °) a pink beam (E=15 keV, $\Delta E/E=10^{-2}$) was used to compensate for the lower photon flux available at a BM beamline. Using a calibrated lithographed Au pattern, the pixel size and the resolution of the FReLoN camera was first determined (0.7 and 1.5 μm respectively); this resolution is good enough to see the desired effects, effects which extend several (~5) μm from the junction edge towards the centre of the shape. Radial scans (reflectivity and large angles, ***Fig. 1***) show the good quality of the metal and oxide layers deposited on the samples (thickness oscillations are visible). The Bragg peak corresponding to the Pt layer (the largest Z) can be identified. Images taken with the 2D detector at different Bragg positions (substrate and Pt) are also shown (***Fig. 1***) – it is fully possible to see the different shapes. Long exposure times were necessary. Attempts to perform the rocking scan approach were unfruitful – the available intensity (even with the use of a pink beam) was not sufficient. The data acquired though allowed to estimate that for a sample containing thicker layers (not necessarily appropriated for tunnel junctions), this approach could be validated (*i.e.* sufficient intensity) at an ID beamline and in pink beam.

While imaging for example the image of a slit in substrate Bragg condition, the sample should act as a mirror. By accurately measuring the image size of the slit (in both directions), it was possible to see, in some conditions and for very thin substrates (i.e. 0.3 mm thick), the sample curvature. The radius of curvature was estimated in both directions of the sample, to ~1.5 and 2.7 meters respectively[1]. Flat samples (1 mm thick substrates) were also available and investigated.

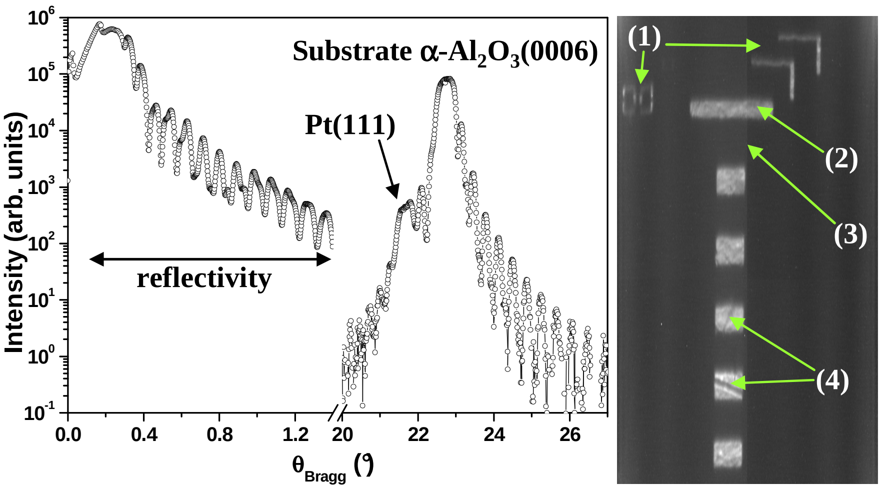

Figure 1: (left) Radial scan (**q** is perpendicular to the surface of the sample, θ-2θ geometry) performed with a point detector. The horizontal axis is labeled as the incident angle. The increased width of the substrate peak is due to the energy spread; (right) Images of the junction area taken at Bragg position corresponding to the Pt peak (sum of 16 images of 30'' each). The image was corrected in the vertical direction (projection by the incident angle θ=21.6°). (1)=mask alignment marks for lithography, (2)=contact electrode, (3)=contact area of the FReLoN chips, (4)=square junctions (some of them with defects / scratches). If the same image is taken at the substrate Brag peak position, the sample acts as a mirror and the shadow of the slit is visible (if the slit opening < camera field of view), with uniform intensity.

The intensity problem can be overcome by using more sensitive detectors: scintillators (point detectors). Indeed, combined with slits, the background can be drastically reduced. In that case, the lateral resolution will be only given by the size of the x-ray beam and focused beams have to be used. We also used monochromatic beams (7 keV) – in this case, the broadening due to the energy resolution is limited. We used ~40 Be lenses to obtain a spot size ~3 × 8 μm (fwhm, v × h) at the sample position, with a photon flux of ~10^8 ph/s.

The Bragg peaks were identified as shown in ***Fig. 1***. Due to the presence of the Kissing fringes, it is difficult to separate the contribution from the different layers – quite often the characteristic peaks appear simply as 'shoulders' – see ***Fig. 2***. By comparing measurements performed at various q-values, sensitivity to different structures (crystallinity) is obtained. By tuning the diffractometer's angles such to be sensitive (in q-space) to different structures, the lateral position of the sample is scanned and thus a raster map of the sample is obtained [2]: an example is shown (on purpose on a 'defect' sample showing errors / misalignments of the masks during the lithography process) in ***Fig. 2***.

A 'good' sample (Co/$CoFe_2O_4$) was then mounted, and the characteristic peaks (Co and $CoFe_2O_4$) were identified. In this case, a rocking scan has to be performed at each position on the junction. This time-consuming procedure limited thus the number of investigated samples / shapes. Still, we could measure effects in 2 directions (// and perpendicular to the x-ray beam) on 2 Bragg peaks (Co layer and $CoFe_2O_4$ layer) for 4 junctions (1 disk, 1 square and 2 rectangles): in all, 16 line-cuts were measured as position-angle maps. One example of the measured and extracted data for the Co layer, in the case of a rectangular junction is shown in ***Fig. 3***. We do not only confirm some of the results reported before [1] but it is also possible to:

-perform measurements of tilts in the direction perpendicular to the x-ray beam

-evidence that these effects are also appearing on the long / short edge of the junctions

-show that for a bended sample, these effects (~1-2°) are 1-2 order of magnitude larger than the geometrical bending of the substrate (~0.02-0.05°) and the focused beam divergence (~0.06°)

-evidence and confirm the idea about the size and shape effect [1].

This approach, with improved spatial resolution (smaller x-ray spots) seems to be extremely interesting in studying local defects on the crystalline materials, at a micron scale, defects which can have various origin (in

[1] These values were confirmed after the experiment by optical measurements of the radius of curvature of the sample (1.4±0.1 m and 3±1 m respectively)

this case, the lithography process / presence of a sharp edge / objects shape). In the particular case of the tunnel junction systems, crystalline defects are in general expected to modify the characteristics (magnetic behavior / functioning characteristics) of the junction.

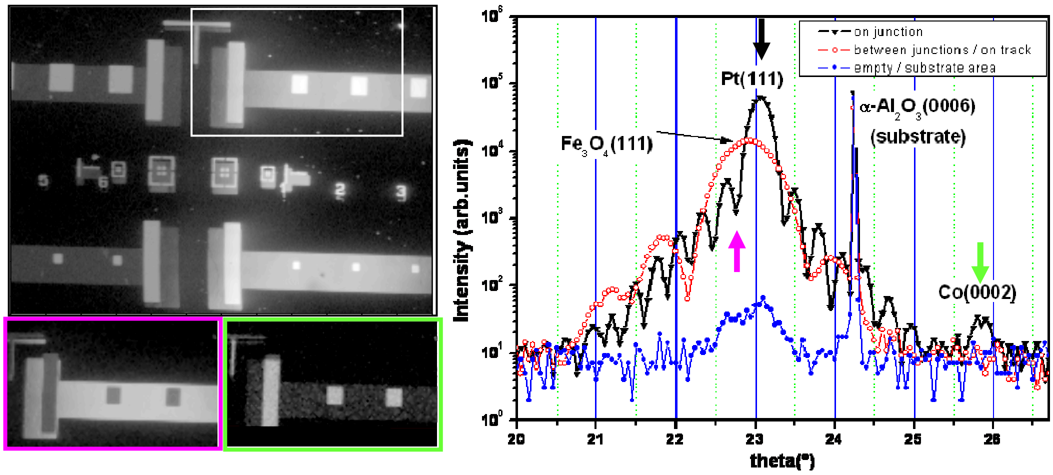

Figure 2: A 'defect' lithographed sample (see text for the details) and of the different constituents (Co/Fe_3O_4) is used. (right) Radial scan (**q** is perpendicular to the surface of the sample, θ-2θ geometry) in the vicinity of the Bragg peaks characteristic to the layers (Pt, Co, Fe_3O_4) performed with a point detector in monochromatic (7 keV) focused beam, at different lateral positions on the sample. Note the change in the period of the thickness fringes (corresponding to a reduction of the thickness) from the junction to the area between junctions on a contact line[2]. Positions in the reciprocal space at which raster maps were performed are marked by the colored arrows. (left) Several raster maps at marked positions (the color of the frame corresponds to the position on the radial scan). Details / alignment marks / defects in masks alignment between processes are visible. Note as well the change in contrast on the objects for the different images.

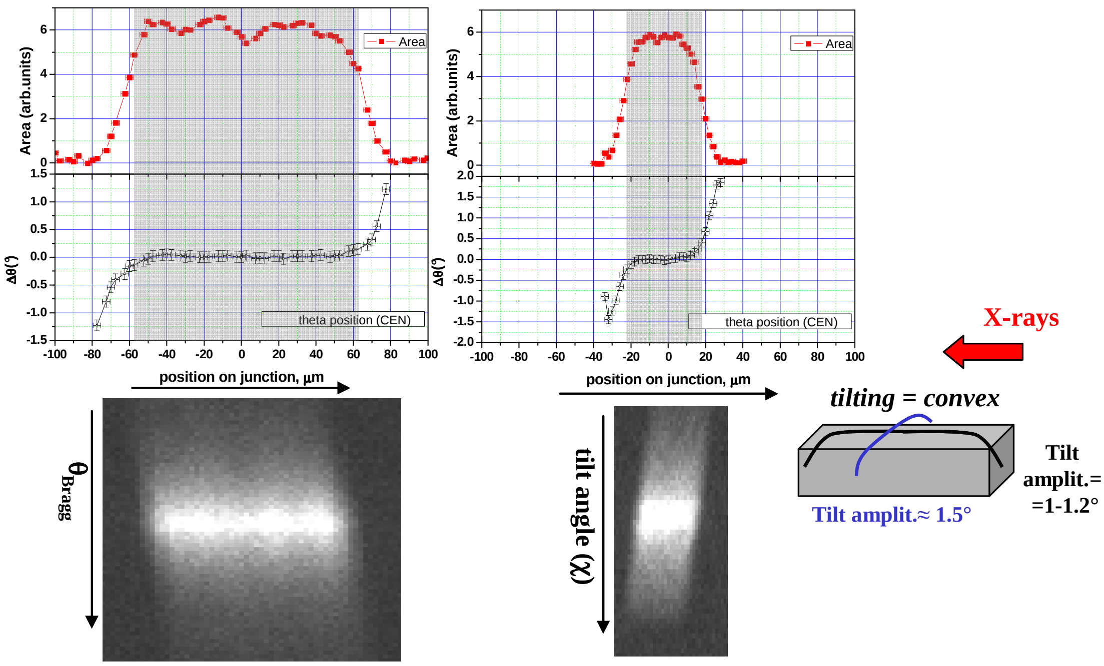

Figure 3: Co plane tilts measured for a rectangular junction (Co/Fe_3O_4 sample), in 2 directions: (bottom) 2d maps lateral position – angle are reported (arb. units for the x and y scales). It can be seen that when approaching the edge of the junction, the center of the peak (angle) is shifting; (top) from the above mentioned maps, the integrated area (describing the junction shape) and the shift of the peak (with respect to the centre of the junction) are extracted at each lateral position on the junction. Although in the automatic procedure this shift is calculated for all data, here is not reported for Area ~0 because is meaningless (it is only a result of 'fitting' noisy background; this is also seen by the noisy behavior of the values found in that region). The error bar represents the angular step used when scanning (typically 0.1-0.2°). A cartoon of the junction orientation and the tilts of the crystalline planes is also shown.

[1] C. Mocuta *et al.*, Appl. Phys. Lett. **91**, 241917 (2007); Eur. Phys. J. Special Topics **168**, 53–58 (2009).
[2] C. Mocuta *et al.*, Phys. Rev. B. **77**, (2008) 245425).

[2] The contact line (Fe_3O_4 layer only) is approximately 2 times thinner than the full junction structure.