ESRF	**Experiment title:** Towards Optimised Spin Seebeck Thermoelectric Generation Devices: by in-situ Pythographic Imaging of Magnetic Domains RE3Fe5O12	**Experiment number**: HC-3502
Beamline: ID01	**Date of experiment**: from: 27 June 2018 to: 3 July 2018	**Date of report**: 19/02/20
Shifts: 18	**Local contact(s)**: Tobias Schulli	*Received at ESRF:*

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The Experiment Setup:

HC-3502 on ID01 was a great success. We have developed a new simultaneous magnetic and structural microscopy based on scanning diffraction, by combining circular polarized X-rays and nanobeam diffraction[1]. The circular polarised Hard X-rays were generated by implementing a diamond phase plate borrowed from the XMaS beamline of the ESRF and resonant X-ray magnetic scattering was set-up by tuning the incident photons to the Gd L2 absorption edge. The X-ray nano-beam was produced by using a Fresnel Zone plate to focus the beam to approximate dimensions of 200nm (V) x 400nm (H). This set-up provided us with a unique scanning nanoBeam magnetic diffraction, which was used to scan over the sample by translating the Fresnel zone plate optics.

The sample Investigated:

The sample investigated was a prototype Spin Seebeck Effect (SSE) device structure, consisting of a thin 23nm epitaxal layer of $Gd_3Fe_5O_{12}$ (GdIG), grown on $Gd_3Ga_5O_{12}$ substrates, with a 10nm Pt layer on the surface. Hall Bars were eteched onto the surface of the film and Pt layer for the measurement of the SSE. Figure 1a-c shows the sample and micrograpgs of the Hall Bars. The shiny areas in fig.1b&c are the film and Pt surfaces the dark areas are the substrate only, where the film is etched away. Normal scanning nanobeam diffraction images of the sample are shown in Fig. 1d&e taken on the (008) diffraction peak of GdIG. The orange are the high diffracted intensity of the film, the blue are zero intensity where the film is etched away. The rocking curve of the film Bragg peaks are around 0.03 degrees, which show the excellent quality of the samples.

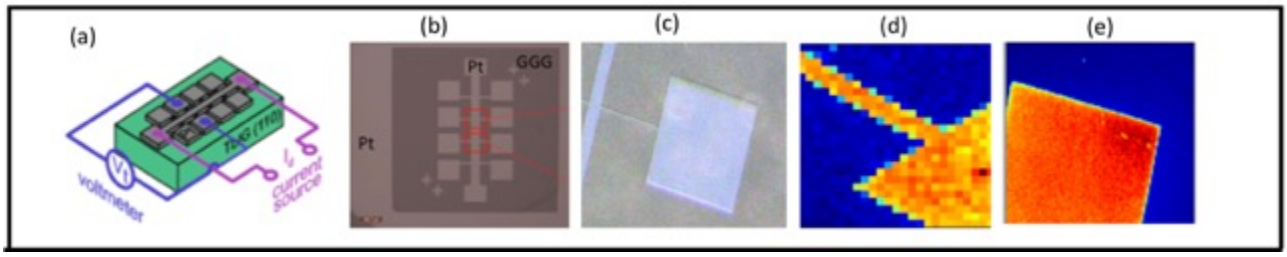

Figure 1. (a-e) The lithography structured sampled investigated consisting of prototype device structure of SSE materials. Hall bars are etched into the sample. (d-e) normal scanning X-ray nanobeam images of the sample, the orange parts are high diffracted intensity from the GdIG film, the blue are zero intensity showing the area where the film is etched away.

The Measurements:

In the experiment at ID01, we used a He flow cryostat to cool the sample to around 5K to maximised the Resonant Magnetic Scattering from the Gd ions from GdIG. GdIG is ferrimagnetic so the (008) Bragg reflection investigated contains both structural and magnetic scattering. The magnetic and structural scattering can be separated by using circular polarised X-rays. Magnetic maps can be isolated for from the Bragg peak by determining a magnetic flipping ratio (FR) defined as $FR=(C_L-C_R)/2*(C_L+C_R)$, where C_L (C_R) refer to the scattered intensity for left (right) incident circular polarisation (fig. 2). The FR measures the components of the magnetic moments in the scattering plane. Using the same datasets, the purely structural contrast is extracted from the sum $S= (C_L+C_R)/2$. This allows us to collect image maps for the magnetic domains and crystal nanostructure from the same data simultaneously and to uniquely correlate how the crystal nanostructure effects the magnetic one. We exploited this new microscopy method to correlate the magnetic and crystalline textures of prototype device structures of the Spin Seebeck thermoelectric compound $Gd_3Fe_5O_{12}$ (GdIG). The insight into the magnetisation so far obtained in these magnetic nanobeam mapping experiments has a spatial resolution limited ~400nm. The results are shown in figure 2.

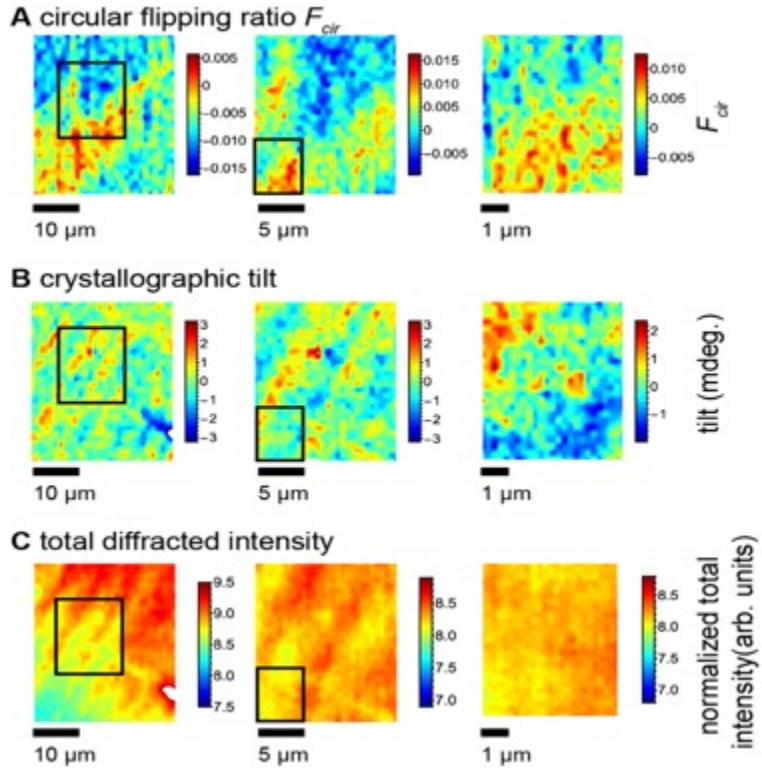

Figure 2: Interaction between Crystallographic and Magnetic Microstructure. (A) Nanobeam diffraction maps of FR at micrometer length-scales. (B) Crystalographic tilt towards the (100) direction. (C) Integrated diffracted intensity at the (008) Bragg reflection. The magnetic response to the structural variation in B&C competes with the development of facets along directions of the lowest domain boundary energy.

The magnetic sensitivity of the microscopy developed depends crucially on the interference of the magnetic and structural (charge) scattering amplitudes. This is possible with the circular light used and provides sensitivity to the magnetic moments in the (vertical) scattering plane used. It is also possible to have this magnetic-structural interference with π-polarised linear X-rays [2]. However, in

this case, the magnetic sensitivity is for the magnetisation perpendicular to the scattering plane, i.e. orthogonal to the sensitivity obtained with circular polarised X-rays. The magnetic maps for circular and π-polarised linear X-rays are shown in figure 3. These results confirm the potential power of the new magnetic microscopy.

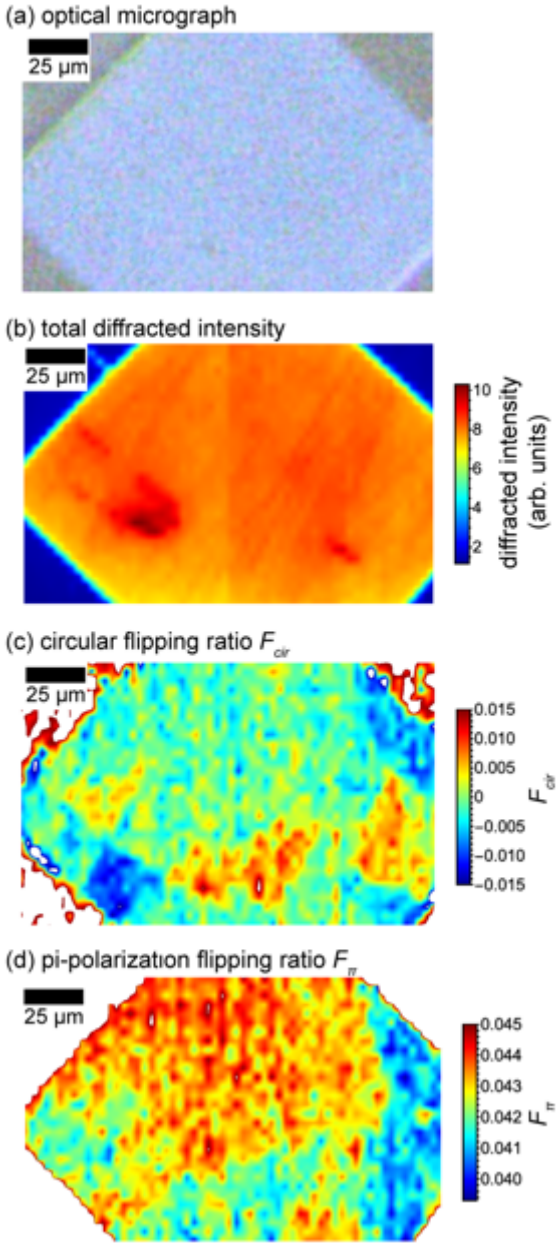

Figure 3. (a) Optical micrograph of part of the sample and (b) normal Bragg scattering of the (008) scaning diffraction maps. (c) shows the magnetic maps obtained using circular polarised X-rays, sensitive to magnerisation in the scattering plane and (d) the magnetic maps obrained using incident linear π-polarised linear X-rays, sensitive to magnetisation perpendicular to the scattering plane.

Conclusion and Future Directions:

We have developed a new tool for the simultaneous investigation of magnetic and crystalline nanostructure which has the potential to address many issues in the development of new spintronics devices. the design and understanding of future spintronic nanotechnologies are severely limited by the lack of characterisations tools of 3D magnetic nanostructure structure. In particular, when crystalline materials are synthesised by epitaxy, the understanding of how the magnetic properties become intertwined with crystal structure and strain at the nanoscale is required. There is little experimental or theoretical insight available into the complex magnetic configuration that can be expected in nanoscale layers in zero magnetic field. In thin films, the magnetization pattern could be expected to arise from a competition between effects arising from nanoscale geometry, elastic distortion, and magnetic anisotropy. The anisotropy problem is important because the SSE depend

sensitively on the magnetization direction in thin layers near the magnetic insulator/heavy metal interface. While magnetic imaging can be performed by a variety of methods including PEEM, this does not provide the correlation of magnetism and nanocrystalline structure of our microscopy. Furthermore we have demonstrated the posibility of imaging magnetisation in two directions in and perpendicular to the scattering plane and penetrating through the Pt surface electrode.

We now wish to extend this microscopy to 3D Bragg ptychography methods in collaboration with experts in this field in a new proposal to ID01. This proposed experiment will take full advantaged of the new ESRF EBS upgrade, where the increase in coherent flux will be vital to exploit the weak magnetic scattering in ptychography imaging.

Our results todate have been submitted to Science Advances and is currently under review and we expect publication very soon.

References

[1] D. Haskel et al. *Europ. Phys. J. Spec. Top.* **208**, 141-155 (2012)

[2] Evans, P. G.; Marks, S. D.; Geprägs, S.; Dietlein, M.; Joly, Y.; Dai, M.; Hu, J.; Bouchenoire, L.; Thompson, P. B. J.; Schülli, T. U.; Richard, M.-I.; Gross, R.; Carbone, D.; Mannix, D. Resonant Nanodiffraction X-ray Imaging Reveals Role of Magnetic Domains in Spin Caloritronics. *submitted to Science Advances* **2020**.