ESRF	**Experiment title:** Magnetic Coherent Diffraction Imaging with hard X-rays	**Experiment number**: MI-1352
Beamline: ID01	**Date of experiment**: from: 09.12.2020 to: 13.12.2020	**Date of report**: 07.03.2021
Shifts: 12	**Local contact(s)**: Steven LEAKE	*Received at ESRF:*

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Report:

The proposal was submitted as a Long Term Project, which was rejected. Nevertheless, 9 shifts were allocated on ID01 to make a start.
The general purpose is to develop Magnetic Coherent Diffraction Imaging, and as far as ID01 is concerned in this project, mainly in Bragg geometry for antiferromagnets. The project would not be possible without the EBS, because magnetic scattering and Bragg CDI are both photon hungry techniques.

Commissioning of the polariser

Refering to the initial plan of the LTP, it was planned to dedicate the 1st run to establish X-ray magnetic scattering and low temperature nanodiffraction on ID01. However, as we already had the opportunity to do it 2 months earlier during beamtime HC-4054, we decided to focus on the commisioning of the new polariser (developed by the ESRF in partnership with SIMaP) which will be used to obtain circular polarisation and measure circular dichroism.

The polariser is a diamond crystal used as a quarter-wave plate: when hitting its $(2\bar{2}0)$ reflection at 45° from the horizontal plane, the transmitted beam sees a significant birefringence between its horizontal polarisation component (the natural polarisation of the incident beam) andits vertical component. The phase shift can be continuously tuned across the Bragg reflection, allowing to obtain full vertical polarisation from the incident horizontal polarisation, and any elliptical polarisation including circular polarisation, either left- or right-handed.

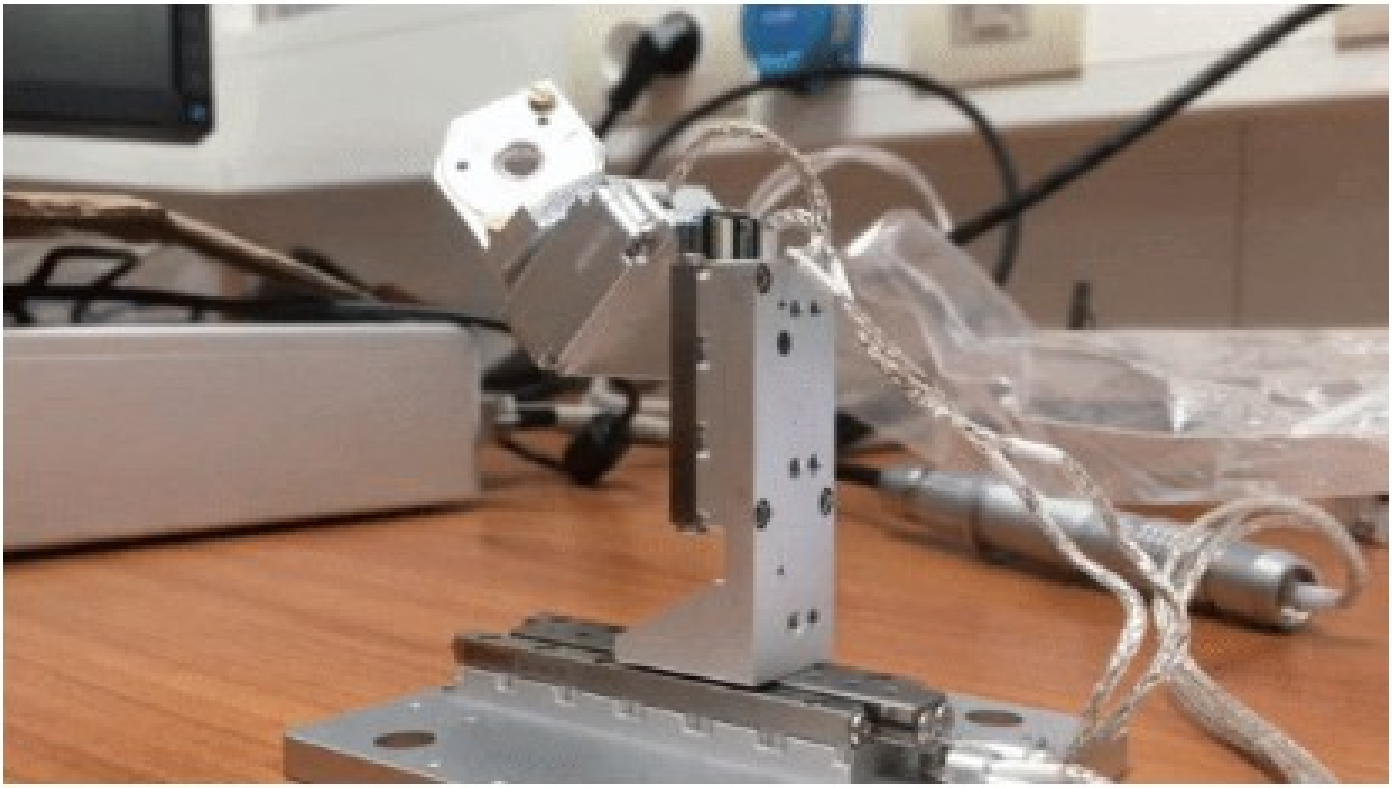

Figure 1: Our recently built X-ray phase retarder used to obtain circular polarisation.

Our polariser assemblee is a stack for SmaarAct motors (2 translations and 1 rotation axis at 45° from the horizontal plane). A diode placed behind the polariser measured the air-scattered intensity at 90° horizontally, which is proportional to the vertical component of the polarisation. A first attempt to align the polariser placed behind the focusing optics (Compound Refractive Lenses, CRLs) did not provide the typical “Batman” scan because of the strong divergence of the beam. After placing the polariser before the CRLs, the expected curve could be observed in the diode at 90°.

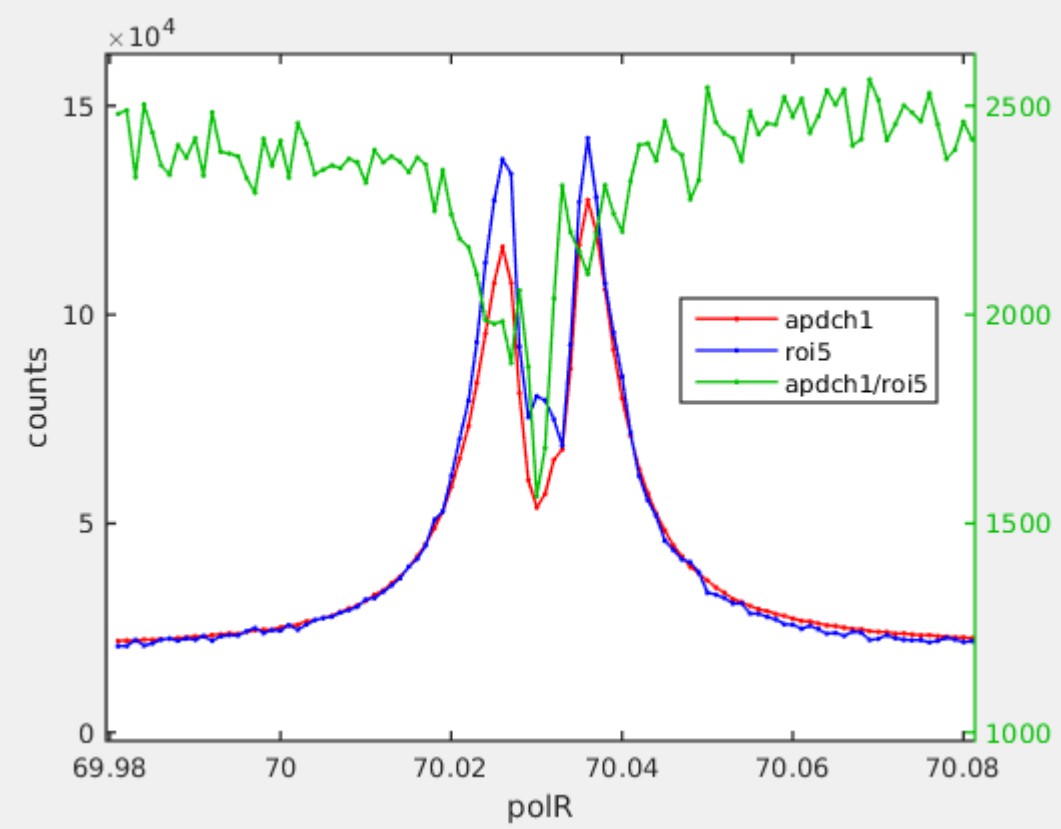

Figure 2: typical "Batman" curves when scanning the polariser angle across the diamond (2$\bar{2}$0) reflection. Red: air-scattered intenesity at 90° horizontally; green: transmitted beam; blue: air-scattered beam normalised by transmitted beam. The maximum of vertical polarisation is achieved on top of the two peaks (blue curve), while circular polarisation is achieved at an angular distance from the centre of the peaks equal to the separation of the peaks. Going from one side to the other reverses the sign of circular polarisation.

We encountered positioning errors on the polariser rotation until we reduced its speed drastically (well below the specifications). After that, we found it very stable.

Gadolinium flake

A macroscopic flake of pure Gd was used to calibrate the energy of the X-rays and find the L2 resonant edge (7.94 keV).

The "Batman" curves of the polariser were measured as a function of incident X-ray energy across the L2 edge of Gd. This procedure allowed to calibrate the shift of the Batman peaks with energy, allowing later to perform simple energy scans with a combined polariser move. This calibration was also the opportunity to observe multiple diffraction below the Gd L2 edge, and to realise that the polariser would be of no use in the energy range 7.92-7.93 keV.

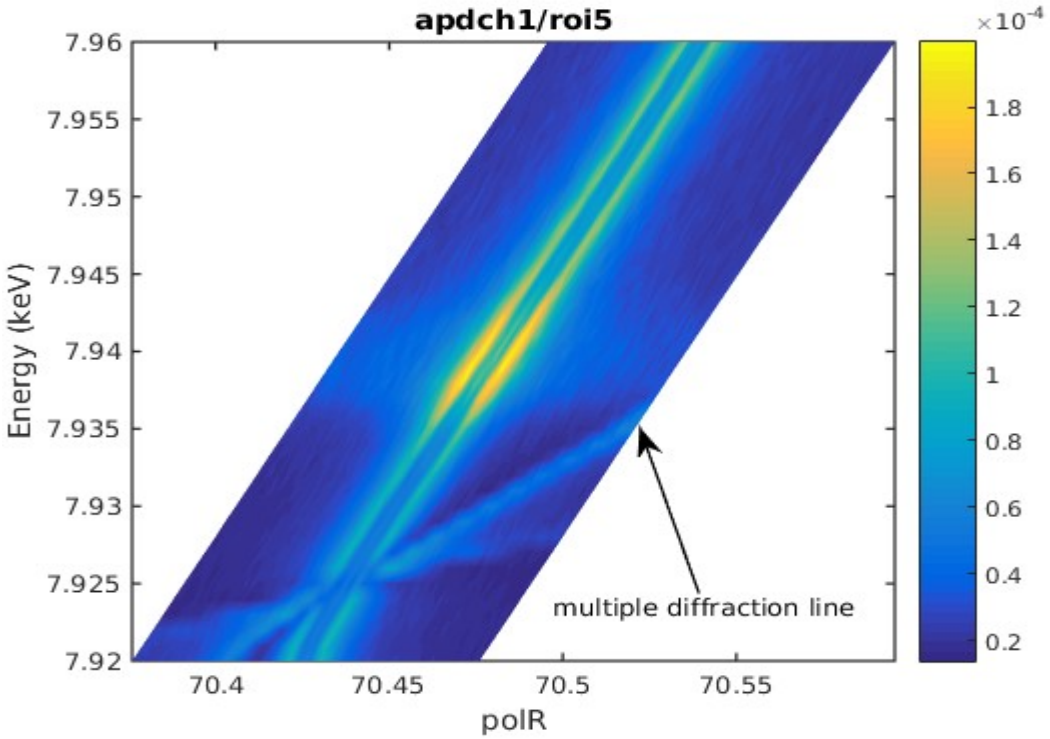

Figure 3: Polariser angle - energy map across the L2 edge of Gd. The resonant edge is seen at 7.94 keV, and multiple diffraction is observed in the 7.92-7.93 keV range.

No XMCD could be measured from the Gd flake, although we cooled it down to 270 K (well below the Curie temperature), probably because it is a soft magnet and has a vanishing net magnetisation (no remanance) without applied field.

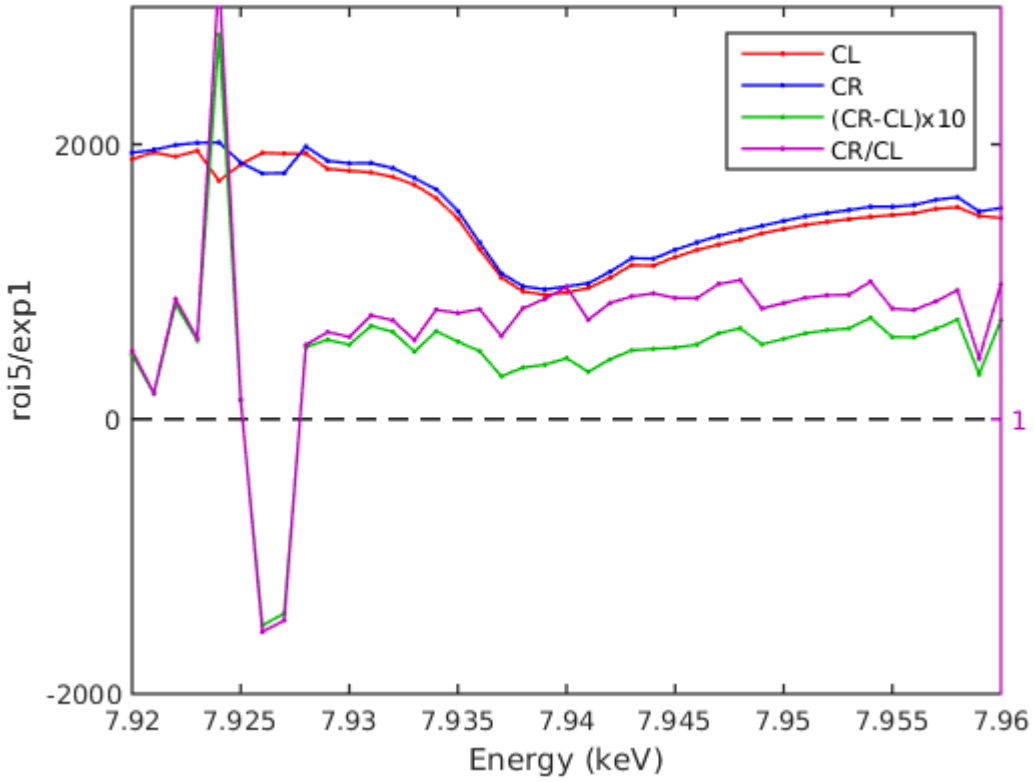

Figure 4: XMCD signal across the Gd L2 edge from the Gd flake. The strong signal in the lower energy range is an artefact from multiple diffraction. There is otherwise no XMCD signal above noise level. Polarisation: CL = circular left; CR = circular right.

Fe/Gd multilayer

A 200 nm thick Fe/Gd multilayer, deposited on a SiN membrane, was mounted on the sample stage in order to look for some XMCD signal. At room temperature and in the absence of external magnetic field, the multilayer host a maze of interlaced domains with typical width of ~150 nm. These were expected to scatter a powder ring at small angles.

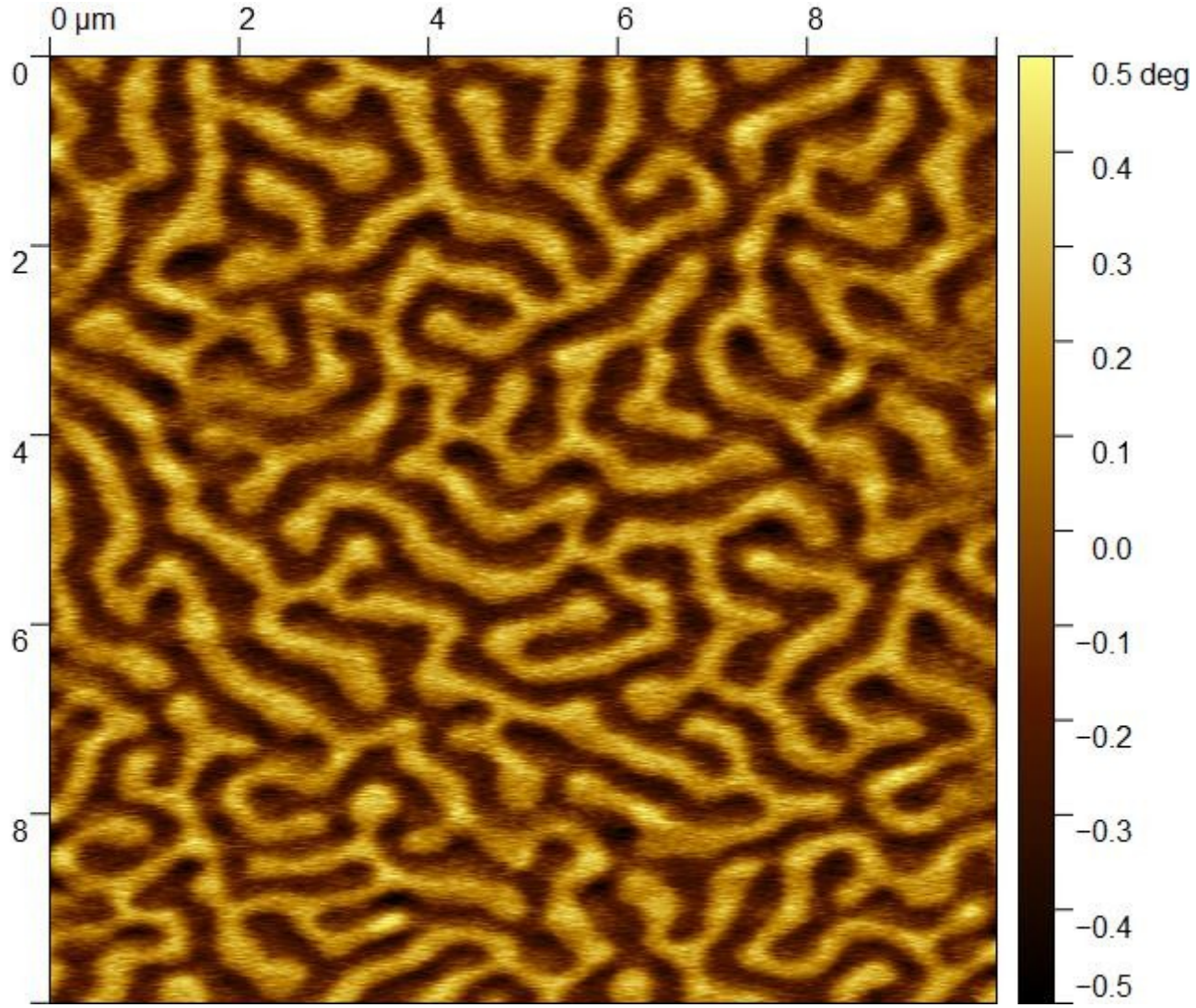

Figure 5: Magnetic force microscopy image of the same Fe/Gd magnetic multilayer deposited on a Si substrate.

This attempt was unsuccessful, as the multilayer was too thin (200 nm) and hence contained to little Gd. We could not even see the absorption edge.

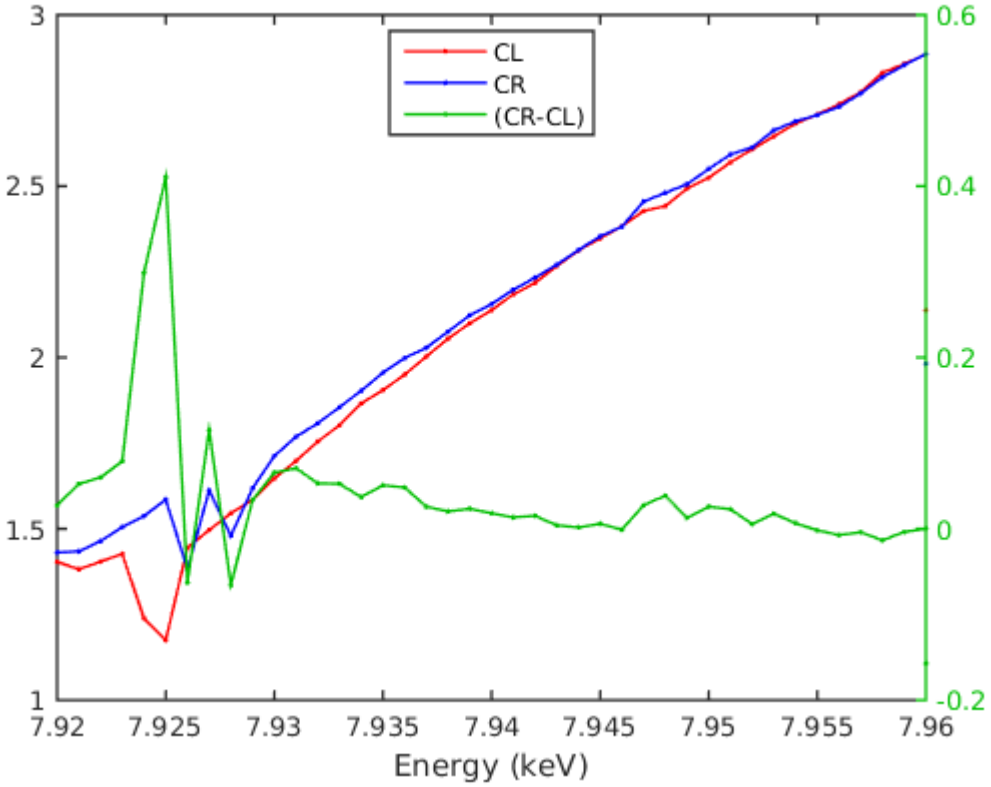

Figure 6: Tentative XMCD across the Gd L2 edge from the Fe/Gd multilayer. The strong signal in the lower energy range is due to multiple diffraction. No significant XMCD signal is seen. Even the absorption edge cannot be seen.

Dy:NdFeB hard permanent magnet

We measured samples of hard magnetic material NdFeB (800 nm thick film) dopped in Dy by annealing a 180 nm thick capping layer at 600°C for 10 minutes. EDX caracterisation suggests that the Dy has homogeneously diffused into the NdFeB film and VSM measurements suggest that the Dy follows the magnetic behaviour of Nd, as noo change could be seen between dopped and undopped samples.

The X-ray beam was tuned to the L3 edge of Dy (7.797 keV), which we had calibrated 2 months earlier during HC-4054.

We had 2 samples: one had been saturated by application of a 7T magnetic field; the other one was in the as-deposited magnetic state, presumably with random domains of magnetisation up or down corresponding to the structural grain size, i.e. ~100 nm.

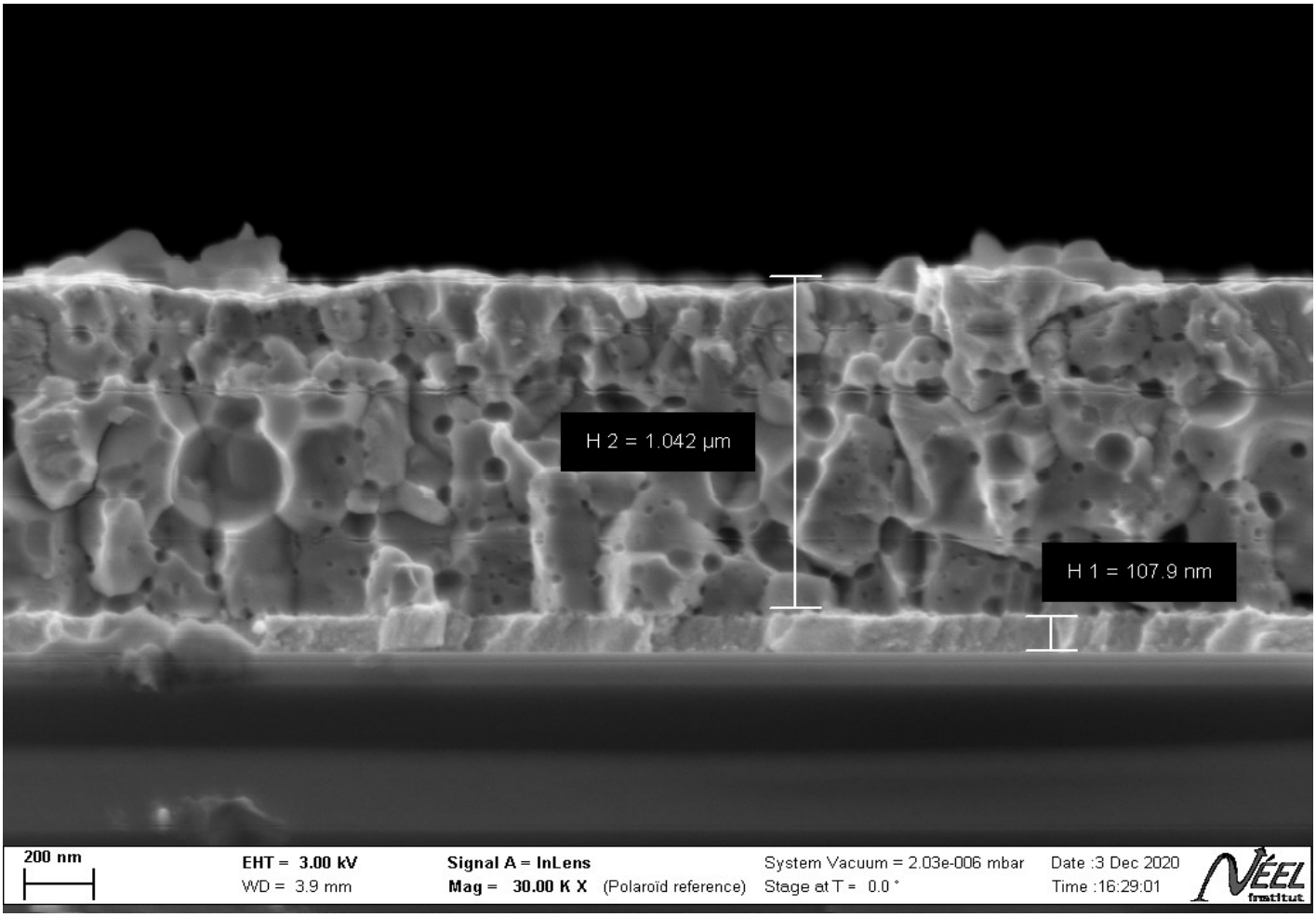

Figure 7: SEM view of the profile of the Dy:NdFeB layer. Note the heterogeneous structure and the grain size of a few 100s nm.

No XMCD could be measured from the saturated sample, and the Dy L3 absorption edge was also not seen in the absorption spectra. Apparently there was not enough Dy in the sample, which is surprising because it is an equivalent thickness of 180 nm.

Nevertheless, we tried with the "as-deposited" sample. We mapped a region of the sample with circular left and right polarisations at 7.797 keV (on resonance) and at 7.795 keV (2 eV below). Although it is clear that the set-up was perfectly stable, as could tell from the charge features, the XMCD maps on resonance and 2 eV below are not correlated. We conclude that at least the measurment 2 eV below the resonance is a measurement of the noise. However, the histograms of values make sense for a weak dichroic signal.

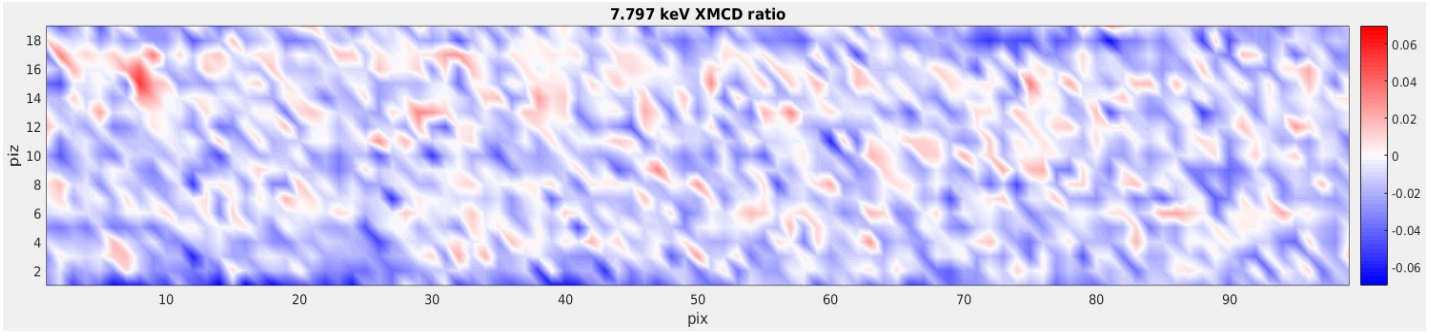

Figure 8: Local map (100 µm x 12 µm) of the XMCD ratio at the L3 edge (7.797 keV). A similar map measured 2 eV below does not show any correlation with this one.

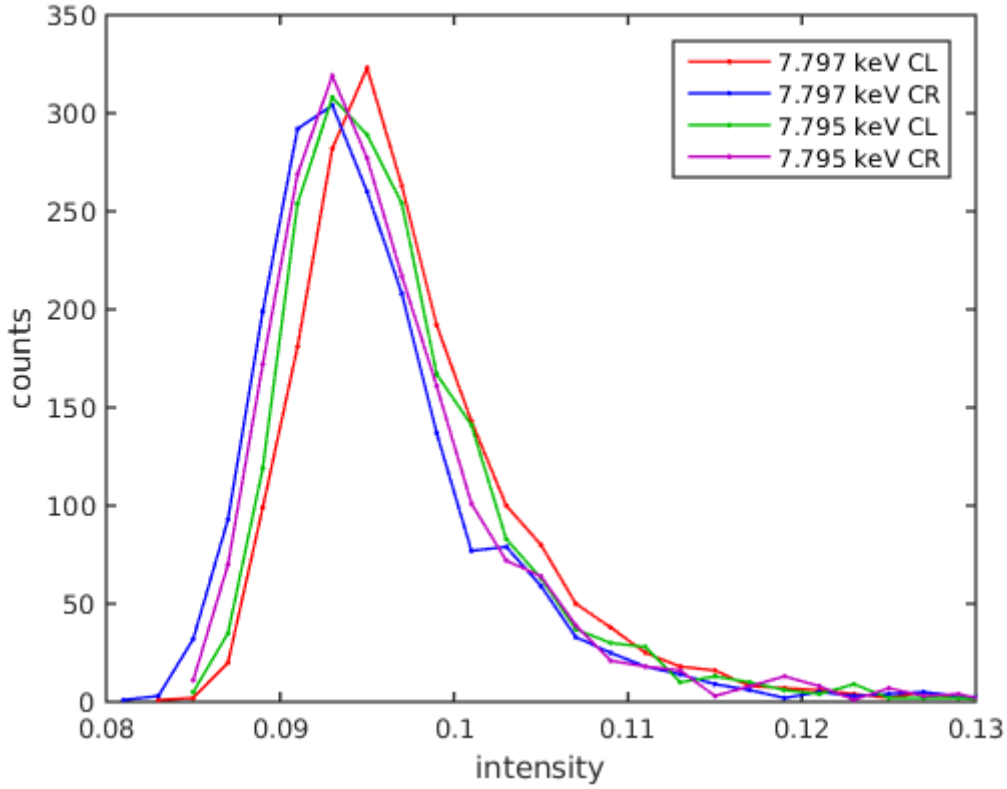

Figure 9: Histograms of values of the maps recorded with circular meft and right polarisations at 7.797 keV and 7.795 keV. The progression of the curves makes perfectly sense for a global dichroic signal.

However it is possible that the X-ray beam (of size 1x1 µm² in these maps) averages over many domains, since the grain size is a few 100s nm. Therefore we used ptychography to achieve better resolution. Several scans were performed and the data is still not fully analysed. We show below an example where dichroic features are visible… magnetic domains? Their shape is suspicious, but fromo the phase retrieval point of view they are reliable.

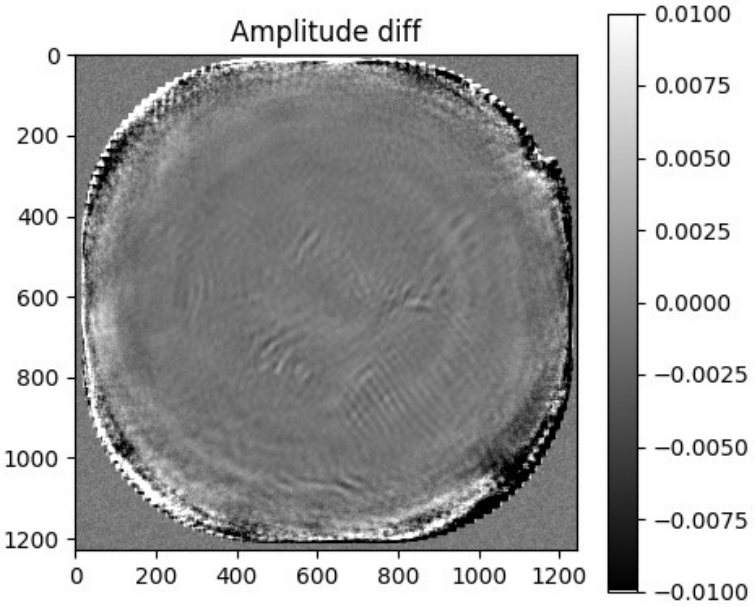

Figure 10: Dichroic image obtained by ptychography. The scale is in nm. Only the central part of the image is reliable.

Dy nanostructures

Nanopillars were etched by Focused Ion Beam (FIB) in a Dy thin film.

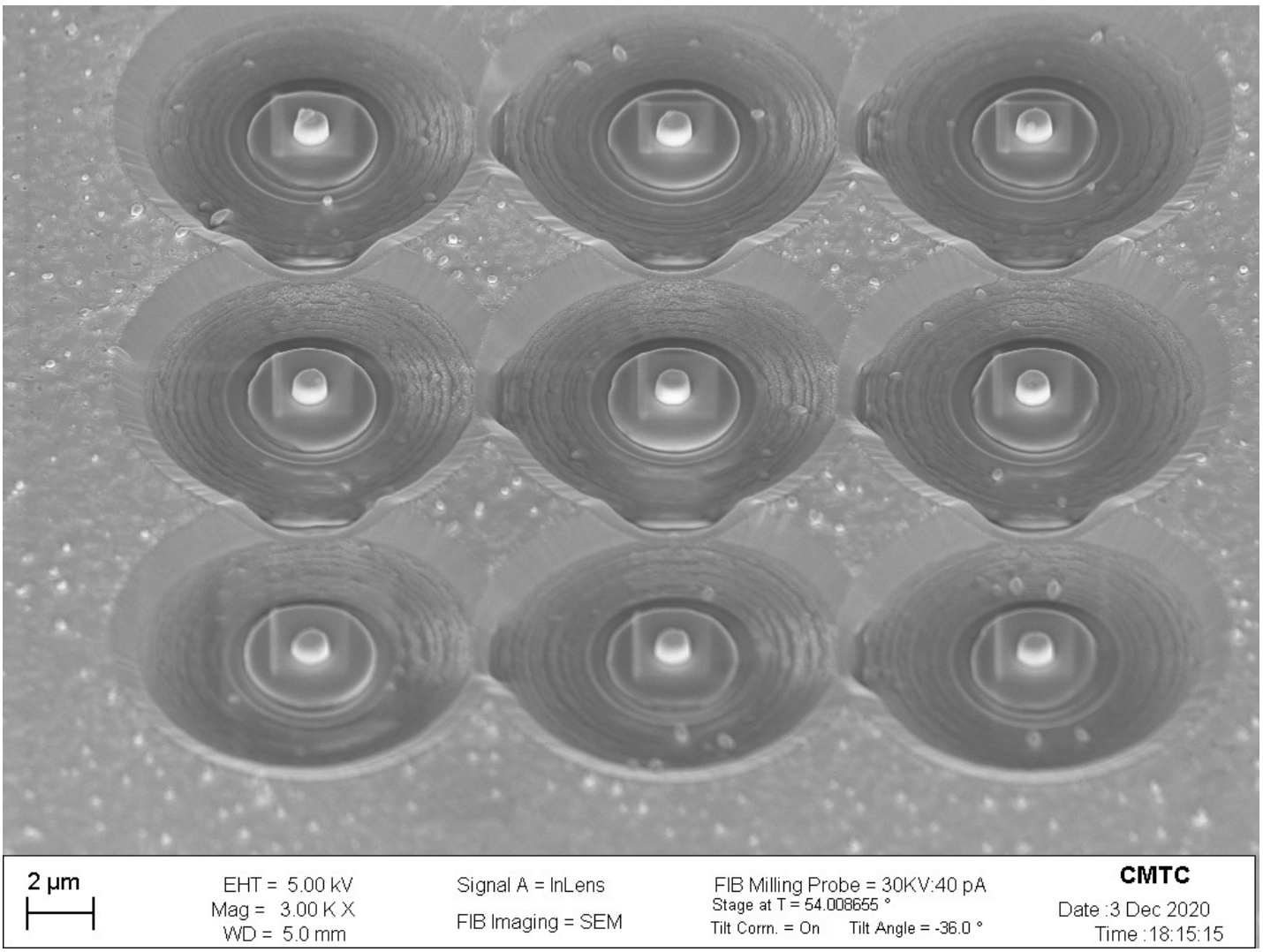

Figure 11: SEM view of the 3x3 array of nanopillars etched in Dy.

We used a similar set-up as for HC-4054 to look for the pure magnetic reflections due to the magnetic helicoidal order between ~90 K and ~180 K: Nitrogen cryostream and polarisation analysis.

The nanopillars could be located by Scanning X-ray Diffraction Microscopy on the Dy (0,0,2) reflection. However, when switching the crostream on, vibration and drift prevents from measuring individual nanopillars. We had to forget about the nanopillars and make measurements on the continuous film away from the etched area.

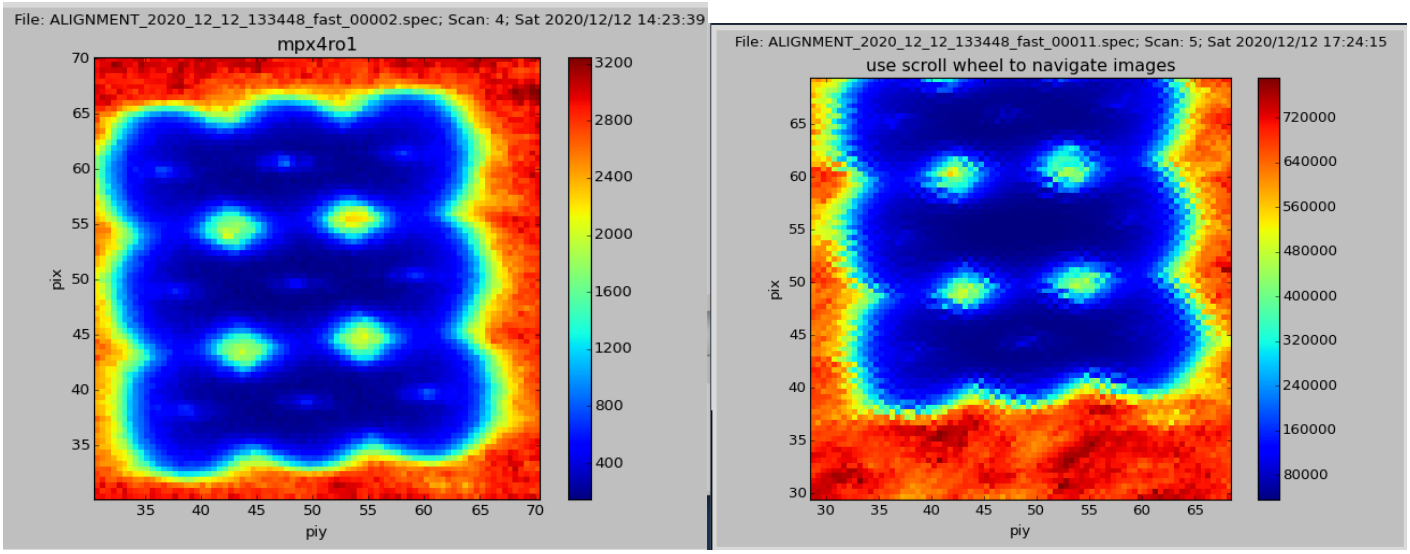

Figure 12: Scanning X-ray diffraction microscopy of the nanopillar array at room temperature (left). The nanopillars are the tiny dots in the centre of the etched disks. When the cryostream is switched on, vibration and drift are visible (right).

The magnetic reflection could not be seen without polarisation analysis, even by using the diamond phase plate to extract the XMCD part.

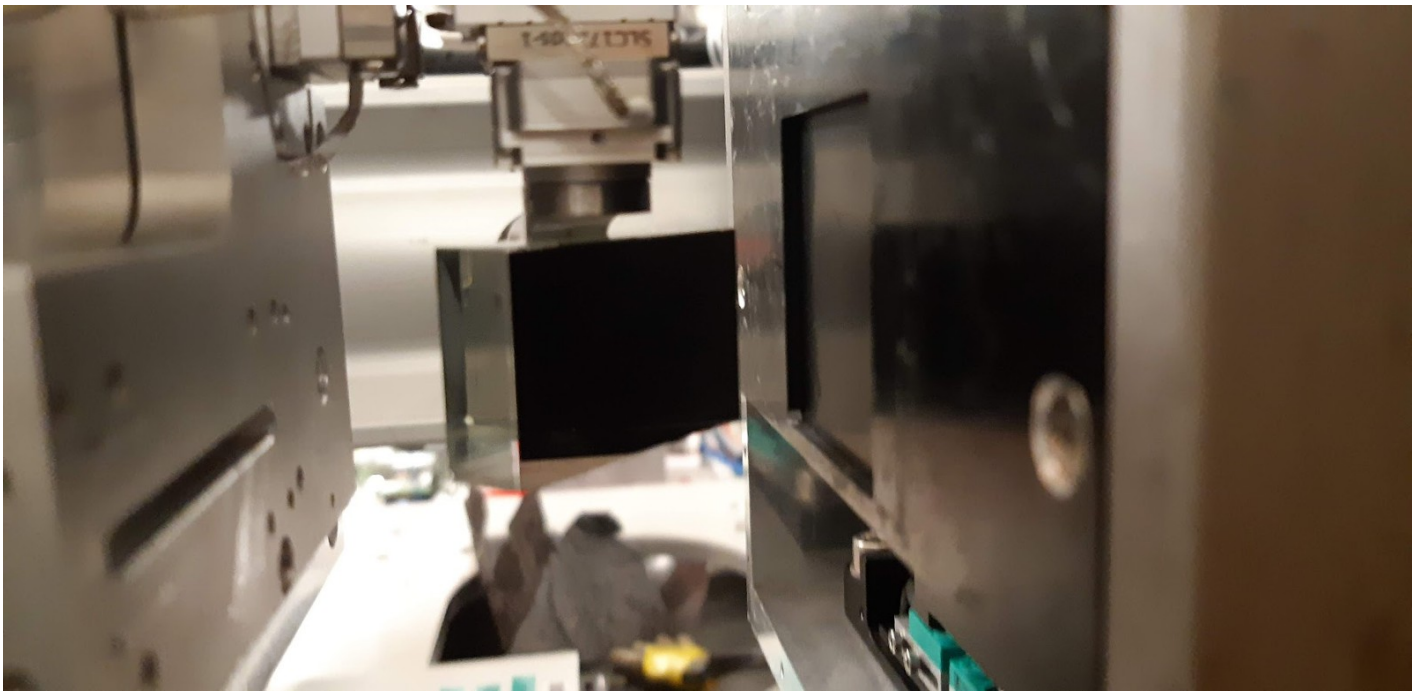

Figure 13: Photo of the bent crystal analyser at 45° incidence for horizontal diffraction at 90°, in front of the detector chip.

Thereefore we installed our polarisation analyser to reject most of the scattered beam of unrotated polarisation. Compared to HC-4054, we had a bent Ge crystal, with radius of curvature 1 m, which we mounted at a distance 1/sqrt(2) from the sample, such that it would in principle allow recording speckle patterns at 90° in a single snapshot, while with a flat crystal we had to integrate over the analyser angle. We tested the idea first on the Dy (0,0,2) charge reflection. The result was promising, but not yet sufficiently good for quantitative measurements of speckle patterns. However for integrated intensity measurements it is much more efficient than the flat crystal analyser.

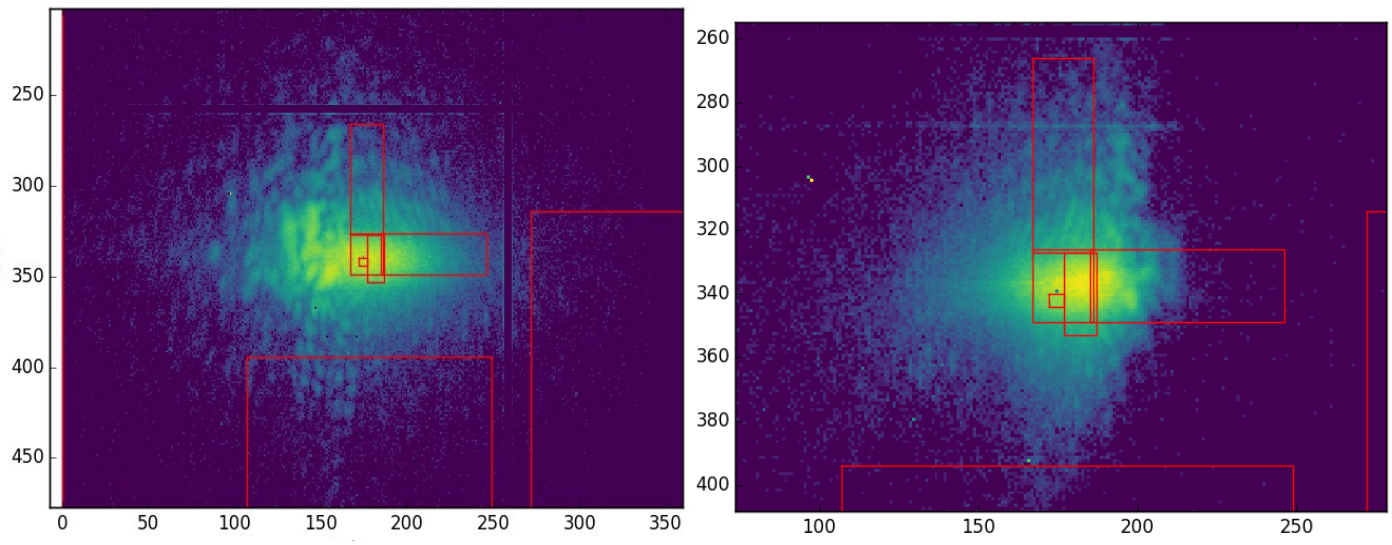

Figure 14: Speckle pattern on the Dy(0,0,2) reflection. Left: without polarisation analyser in 10 seconds. Right: with polarisation analyser in 100 seconds. Note the left-right mirror symmetry due to the extra reflection on the Ge crystal. The speckle pattern is preserved, but with distortion and bluring.

The polarisation analyser allowed to measure the (0,0,2-tau) magnetic reflection at the Dy L3 edge. We tried both circular and linear incident polarisation, in case the circular dichroism would allow to enhance the peak from the beackground. Thsi would happen if the charge amplitude is strong at the position of the magnetic peak. But we found that the linear polarisation was more effficient.

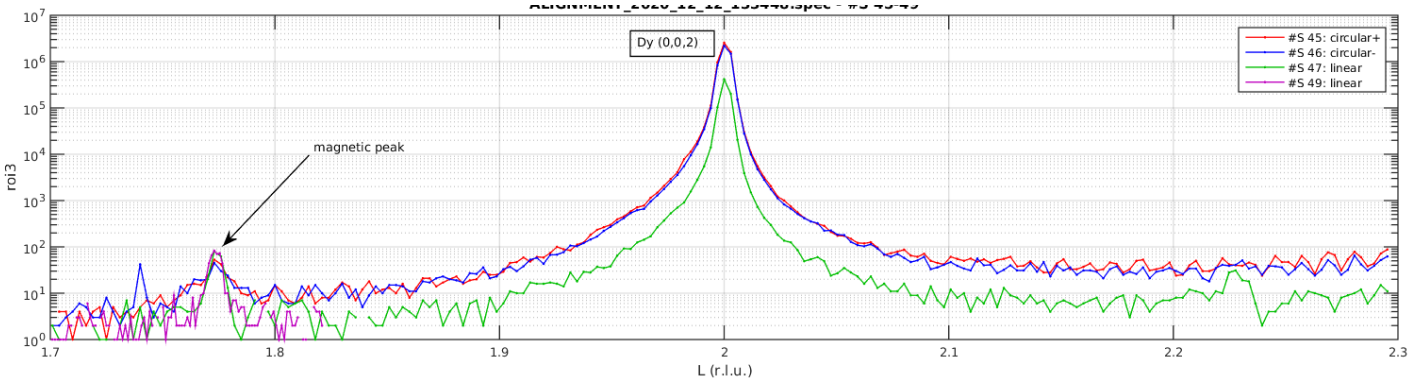

Figure 15: Theta/2-theta scans with circular left and right polarisations and with linear horizontal polarisation, all measured with the polarisation analyser rejecting most of the horizontally polarised X-rays.

Conclusions

We have commissioned the new X-ray polariser. After drastically reducing its speed, its performance is very satisfactory. However, none of the samples we tried dispplayed a clear XMCD signal, while the expected few percent of contrast should be above background.

We have also tried a bent crystal analyser, in order to record speckle pattern in a single snapshot. The results were very promising, but at this stage don't allow quantitative speckle pattern measurements.

The investigation of Dy etched nanostructures showed that for such systems, the XMCD contrast is close to zero and is thus not an efficient way to measure the speckle patterns at the (0,0,2-tau) magnetic reflection. Using incident liear polarisation with a polarisation analysis of the scattered beam is much more efficient.

We also confirmed the difficulties to perform speckle measurements at low temperature, which we already experienced during HC-4054. Some work is needed to improve the stability with the cryostream or to find an alternative vibration-free cooling system.