EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

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 via the User Portal:
https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal ("relevant report")

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *"preliminary report"),*
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as "relevant report(s)" in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

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.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number 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:** A new microscopy of atomic motion in nanostructures	**Experiment number**: MI-1377
Beamline: ID01	**Date of experiment**: from: 17.04.2021 to: 22.04.2021	**Date of report**: 20.09.2021 *Received at ESRF:*
Shifts: 12	**Local contact(s)**: Steven Leake and Ewen Bellec	

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Report:

Summary

The prupose of this experiment was to develop a new microscopy, based on Coherent Diffraction Imaging (CDI) and on the measurement of forbidden crystallographic reflections at a resonance edge. Under such conditions and in the wurtzite crystal structure, the retrieved direct-space image encodes the amplitude of thermal motion instead of the electronic density in the case of conventional Bragg CDI. However the thermal motion induced is not alone and dominates another temperature-independent amplitude only at high temperature [1]. As an interesting application case, we studied GaN nanopillars, which grow with inversion domains [2,3]. Comparing the intensity of thermal motion close to and far from inversion-domain boundaries would be interesting. The effect of the free surfaces of the nanopillars is also interesting.

The experiment was very successful: we managed to record a diffraction pattern at the 115 forbidden reflection of one of the nanopillars, and to retrieve a direct-space image from it. However, the measurement was done at room temperature only, and we did not have time to perform the measurement at high temperature, because we encountered several experimental difficulties.

Experimental details

The beamline was aligned at the Ga K edge (~10.637 keV) and focused with compound-refractive lenses. With coherence slits closed to 0.5 mm x 0.14 mm (V x H), the beam size at the sample was 0.4 µm x 1.1 µm (V x H). The detector was a Maxipix and was mounted at 1.666 m from the sample.

The sample was the same as the one which we previously inversion-domain boundaries [2,3]. It contains a large number of GaN nanopillars grown on a sapphire substrate. Owing to the growth conditions, only few of them have a perfect crystal structure and most of them host inversion domains.

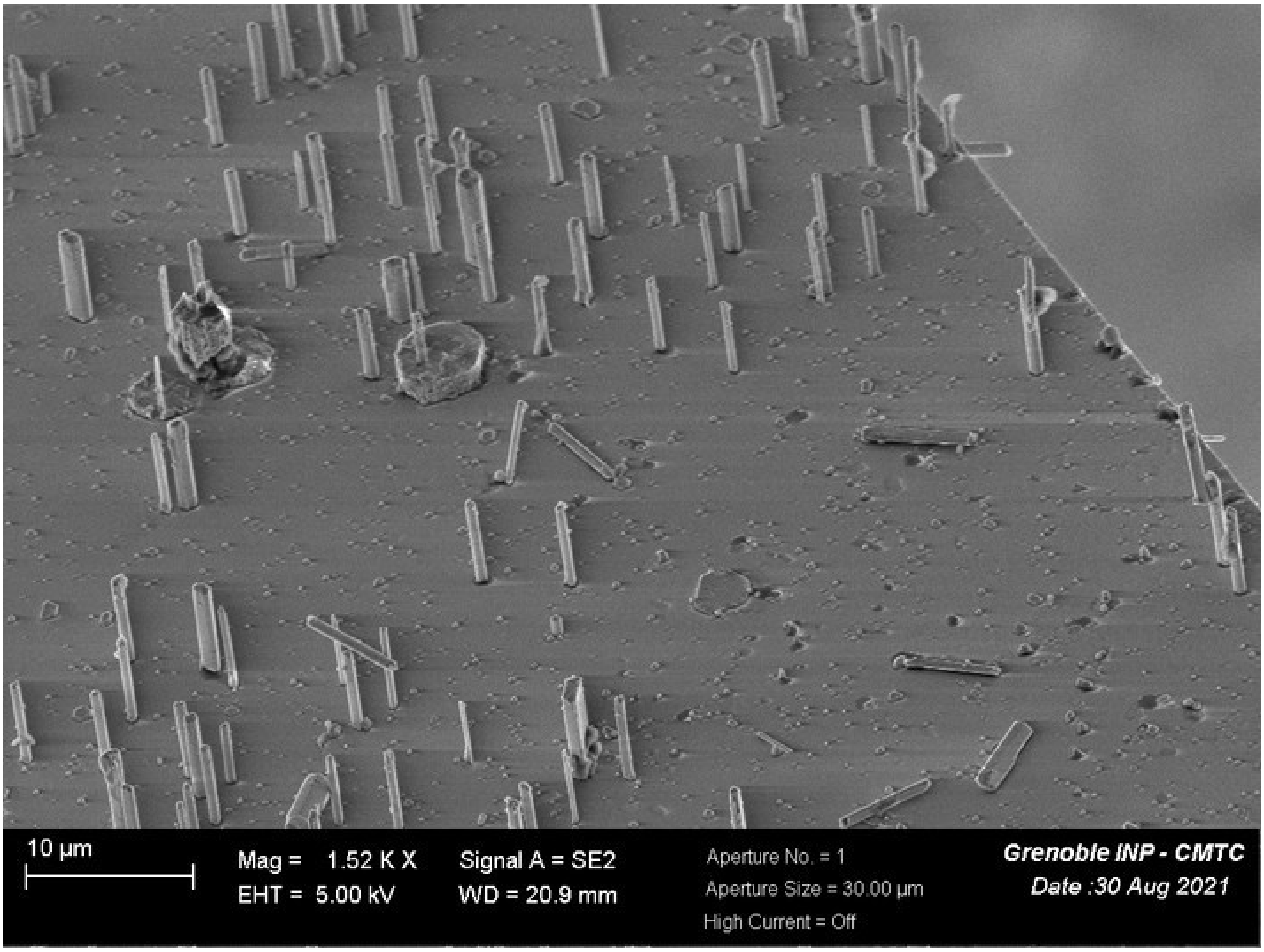

Figure 1: SEM view of a typical area of the sample, close to an edge.

One of the difficulties of the experiment was to find several allowed reflections from the same nanopillar, in order to build an orientation matrix and then look for a forbidden reflection.

We started with the (0,0,4) reflection, which is specular, and then looked for several off-specular reflections among the (1,0,4), (-1,0,4), (1,1,4) and (1,-2,4). Thanks to the diffraction maps, we could make sure that we always measured the same nanopillar. Once we had a satisfying orientation matrix (allowing to find allowed reflections with little effort), we looked for the (1,1,5) forbidden reflection. It could be found in 2 nanopillars. However, automated alignment procedures were unsuccessful, owing to the weakness of the peak, and it could be found only by a careful inspection by eye of the detector images.

Once the (1,1,5) forbidden reflection was found, it was optimised in energy and azimuth, 2 important parameters when measuring forbidden resonant reflections. The energy dependence (Figure 2) was found similar to that in the bulk [1], but with weaker absorption effects. As expected, it vanishes away from the resonance.

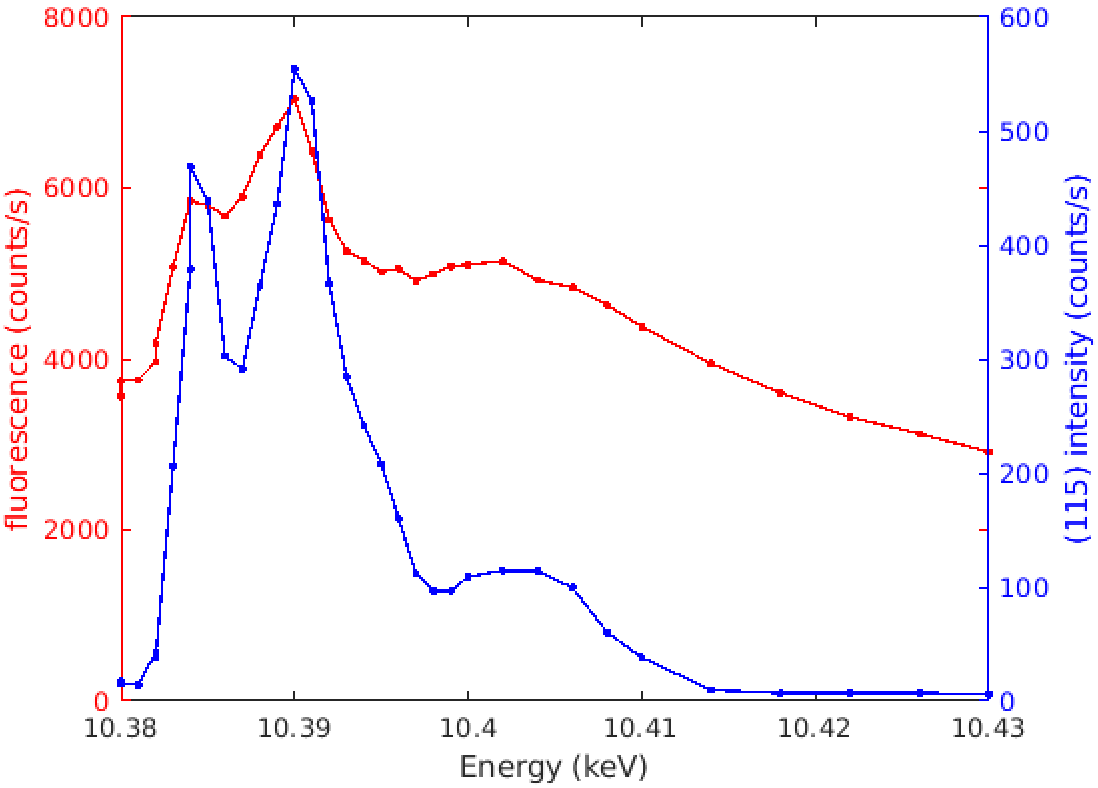

Figure 2: Energy dependence of the (1,1,5) forbidden reflection around the Ga K edge. The fluorescence is also shown for comparison.

The azimuthal dependence (Figure 3) was performed at 10.38 keV, i.e. just below the absorption edge, in order to find multiple scattering events and chose an azimuth sufficiently away from them and therefore being in a situation when dynamical effects can be ignored (a necessary assumption for phase retrieval). Note that we had to develop a routine to perform such scans, by simultaneously scanning phi, eta, delta and nu, since the specifities of ID01 are not covered by the calculations of You. The azimuthal position of the multiple scattering peaks was roughly in agreement with our calculations, although their relative intensities were significantly different. This is due to the fact that the calculations are done for a bulk material. As predicted, the small azimuthal region around 25° was free from multiple scattering contamination and was selected for the subsequent measurements. Moreover, the resonant amplitude also has an azimuthal dependence, which is well known and varies slowly compared to multiple scattering [1], and is nearly at its maximum at 25° (the maximum is at 0° but multiple scattering is extremely dense around this value).

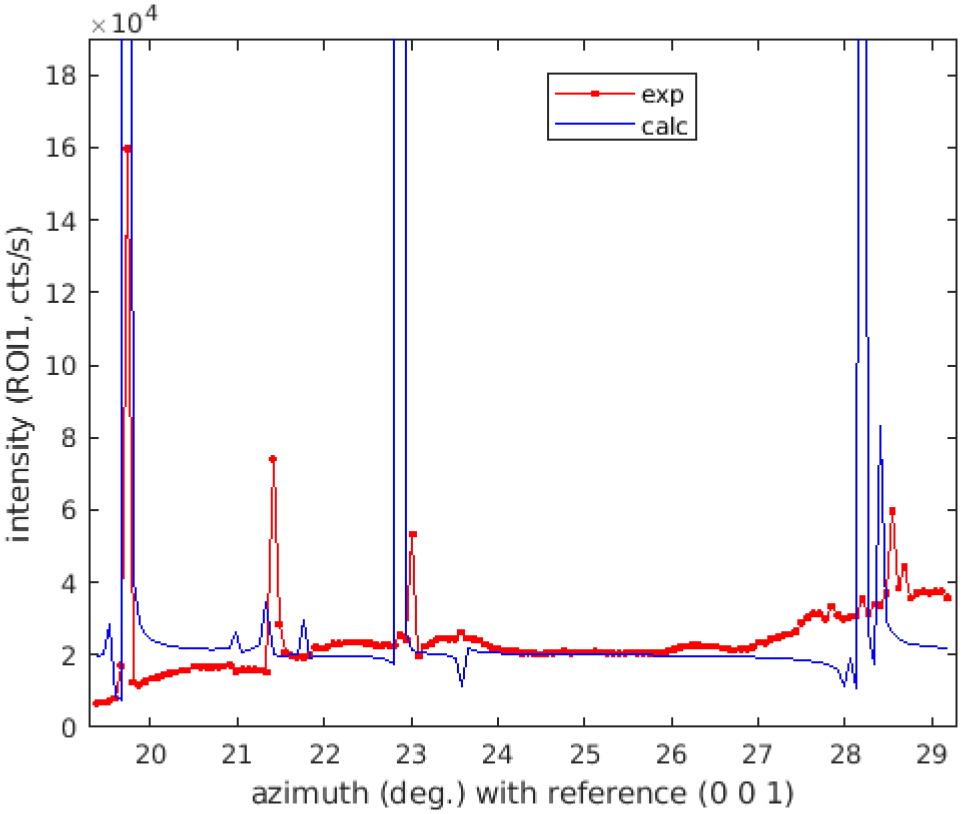

Figure 3: Azimuthal scans on the (1,1,5) forbidden reflection at 10.38 keV. Comparison between experimental data (red) and calculations (blue). The sharp peaks are due to multiple scattering and should be avoided for quantitative measurement of the resonant amplitude.

Radiation damage

After aligning the (1,1,5) forbidden reflection on a first nanopillar, we attempted to record a speckle pattern with high statistics during a night scan. However we found out that the beam severely damaged the nanopillar (Figure 4) and we did a careful investigation of radiation damage before continuing with another pillar.

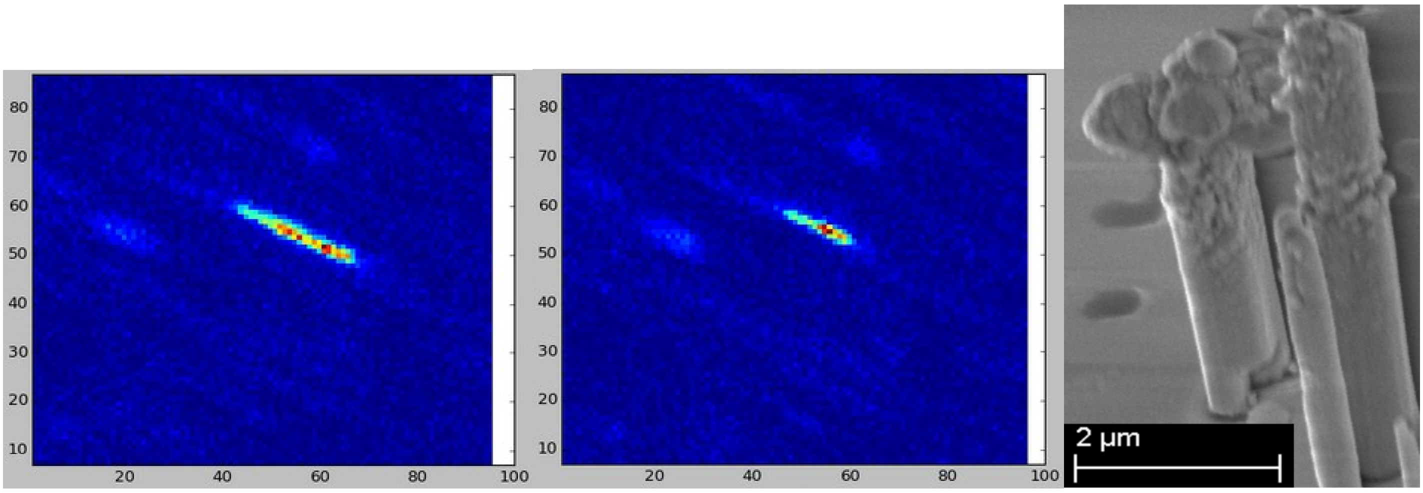

Figure 4: Radiation damage. Left: diffraction map before long exposures. The beam comes from the top left corner. Middle: same diffraction map after long exposures. The bottom right part (i.e. top) of the pillar has disappeared. Right: SEM image of the damaged nanopillar (the one with a "flower" head).

We concluded that exposures of 1 second were acceptable, with a recovery time of ~1s between exposures provided by the deadtime of the control software.

Results

After destruction of the first nanopillar, we had only 24 hours left to complete the experiment. After characterisation of the radiation damage, we aligned another nanopillar. A reasonably good dataset was recorded in one hour (i.e. ½ effective acquisition time) on the (1,1,5) forbidden reflection of a pillar (Figure 5). Unfortunately, we realised only afterward that the (1,1,5) reflection had not been recorded on the same pillar as the (0,0,4) and other allowed reflections. Nevertheles, obtaining a diffraction pattern of sufficient quality for phase retrieval on a forbidden reflection is already a notable experimental achievement.

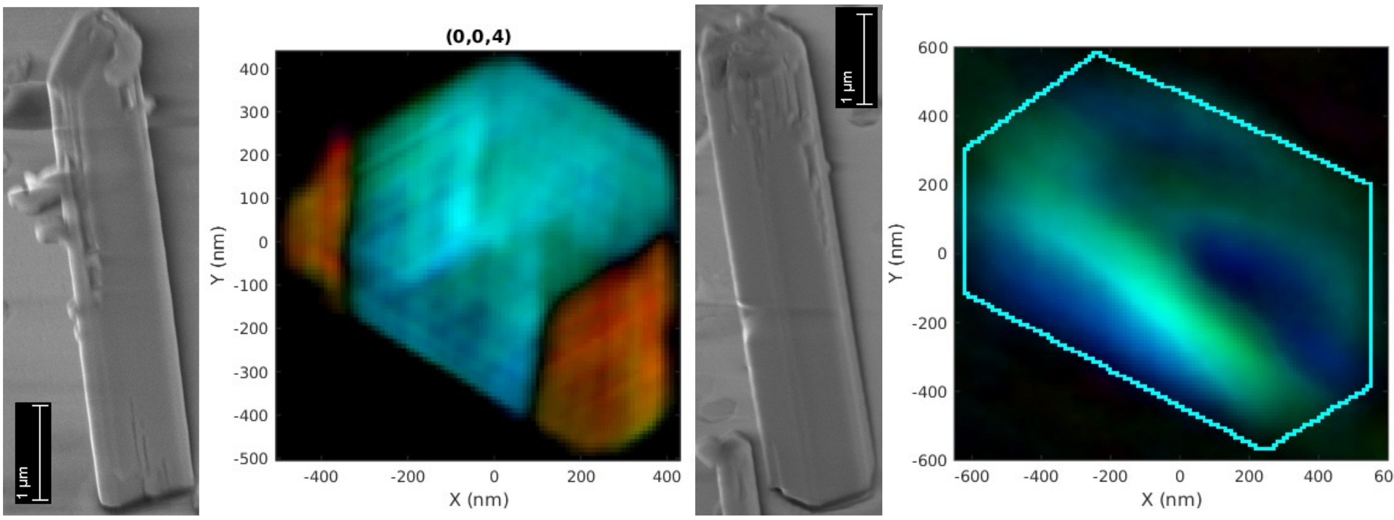

Figure 5: SEM images and direct-space images from phase retrieval of 2 nanopillars: from the (0,0,4) allowed reflection on the left and from the (1,1,5) forbidden reflection on the right. The colours encode the phase and highlight the inversion domains.

After the measurement at room temperature, we started heating the sample and followed manually the (1,1,5) reflection during heating but we lost it. At 500°C, we could find the nanopillar again and measured the (0,0,4) and (1,1,4) allowed reflections. The diffraction patterns at room temperature and 500°C of the (0,0,4) reflection show that the crystal structure was intact (Figure 6). Unfortunately we didn't manage to find the (1,1,5) forbidden reflection again within the little time left.

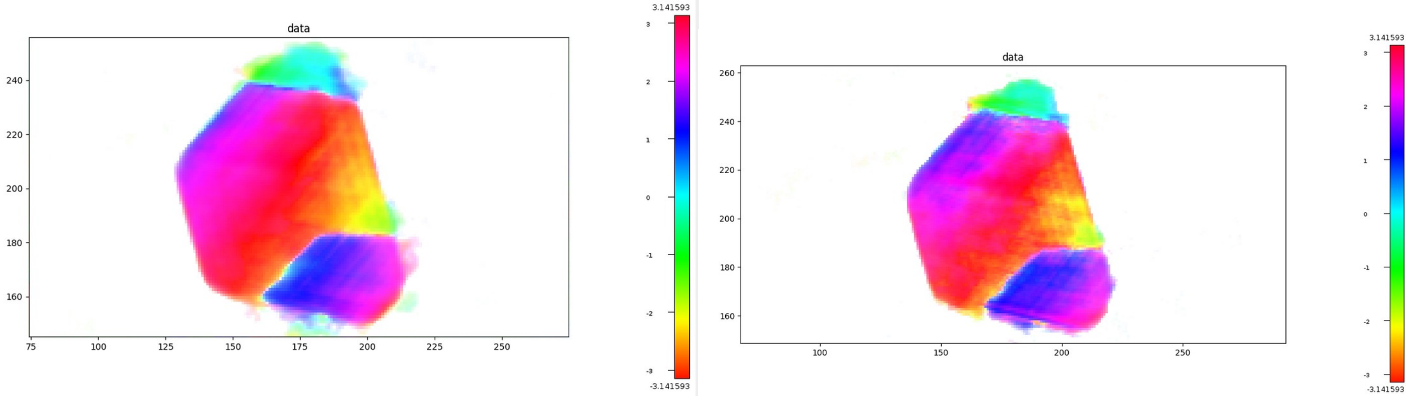

Figure 6: Direct-space image from phase retrieval at the (0,0,4) reflection at room temperature (left) and 500°C (right).

These results are very positive and we are confident that this experiment could be carried out successfully, now that we have optimised our alignment procedures and characterised the radiation damage.

References

1. Beutier et al, Eur. Phys. J. Special Topics 208, 53–66 (2012).
2. Labat et al, ACS Nano 9, 9210-9216 (2015).
3. Ni Li et al, ACS Nano 14, 10305 (2020).
4. You, J. Appl. Cryst. (1999). 32, 614–623 (1999).