Standard Experimental Report

Proposal title: **Looking (again) for magnetic long-range order in infinite layer nickelates**
Proposal number: **HC-5288 (A02-2-889)**
Beamline: **D2AM (ESRF)**
Shifts: **15**
Date(s) of experiment: from: **27 Jun 2023** to: **03 Jul 2023**
Date of report: **05/09/2023**

- Objective & expected results (less than 10 lines): -

Here, we used surface resonant X-ray diffraction (SRXRD) at the Ni K-edge on 10 nm thick infinite-layer nickelate films. To the best of our knowledge, there have been no reports on the use of this technique in very grazing conditions to ascertain the long-range nature of magnetic properties in oxide thin films, such as cuprates, which represent a model system. The detection of a possible peak at a specific (h/2, k/2, l/2) magnetic scattering vector would provide important information about the long-range nature of the previously measured magnetic excitations in $NdNiO_2$ thin films (proposal HC4166).

- Results and the conclusions of the study (main part): -

Infinite-layer nickelates are challenging to synthesize. Contrary to the findings in thin films, superconductivity has not been observed in bulk samples. Indeed, according to the current state-of-the-art the infinite-layer nickelates are believed to not exhibit any magnetic long-range order, since it has not been detected in bulk samples. The latter are likely characterized by a high concentration of impurities which either invalidates or complicates the interpretation of data from techniques requiring a large amount of material, such as neutron diffraction. It is therefore essential to confirm this property in better quality samples, *i.e.* thin films. We proposed the first X-ray magnetic scattering experiments at the Ni K-edge on high quality infinite-layer nickelates thin films, aiming at identifying the presumed magnetic order scattering wave vectors (½, ½, ½). However, due to the profound penetration depth of hard X-rays, thin films are not ideal for such measurements. Our proposed experiment was primarily exploratory, necessitating a highly grazing geometry condition which complicated further the detection of measurable magnetic peaks.
To achieve the very grazing condition ($\alpha < 1°$) necessary to reduce the background signal primarily from the substrate, we used the multiple degrees of freedoms of the goniometer at D2AM. Essentially, for each magnetic scattering plane of interest, *i.e.* (h/2, k/2, l/2) where h, k, l are odd numbers, we selected the one that was accessible at a specific azimuthal angle (ϕ) for which the incidence angle, α, was less than one degree. Our procedure was as follows: before measuring through the analyser in the rotated channel (sigma-pi), we first aligned the detector to the 'charge' scattering peak (sigma-sigma channel, without the analyser) at the chosen 2*(h/2, k/2, l/2) scattering vector position. We then optimized the omegaa-angle of the analyser since there was always a minor 'charge' signal 'leaking' in the magnetic diffraction condition, although with a count rate diminished of a factor around 50. Lastly, the goniometer was adjusted to the specified magnetic scattering condition, *i.e.* (h/2, k/2, l/2). Transverse scans are obtained by rocking the sample (theta-scan) and each scan was repeated multiple times to enhance the signal-to-noise ratio. Overall, the entire procedure took about ten hours for each scattering condition. We were only able to measure a few magnetic scattering vectors, such as

(½, ½, l/2) with l values of 1, 3, and 5. Figure 1-a displays the summed detector images from multiple scans at specific θ value and a transverse scan at 20 K at the (½, ½, ½) Bragg point. A peak is observed in the detector region where magnetic scattered signals are expected. By performing a median operation in the specific ROI indicated by the red box, it is seen that at 20K the result appears as a peak-like which has maximum magnitude at θ=1.1° as shown in Figure 1-b. Such a peak is not present at room temperature, and there's no θ dependency, as depicted in Figures 2a and 2b. We believe the peak is related to long-range magnetic order in the infinite layer nicklates as we've repeated the measurement several times and we still find the same signal.

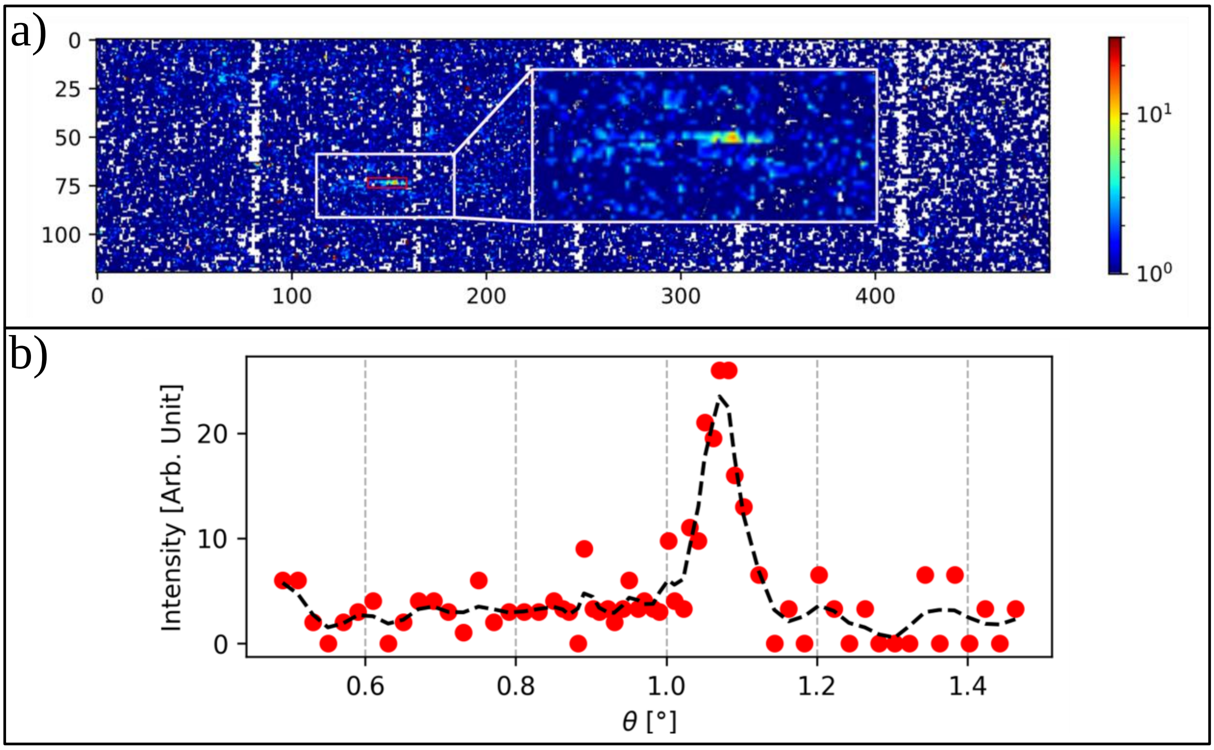

Figure 1: Detector image and transverse scan of scattered X-ray at 20K and (½, ½, ½) Bragg point. a) Summation of all the detector images at θ=1.1° in false color. b) Transverse scan obtained by rocking the sample, the data is obtained from the integral sum of the red ROI in the detector image. Please notes that the resulting transverse scan data is obtained by repeating the same scan for five times then taking median operation of the scans. The dashed black curve is 1D Gaussian filter smoothing of the experimental results.

Additionally, as depicted in Figure 3, no peaks from scattered X-rays are detected on the 2D detector for the scattering vector (½, ½, 3/2) at 20 K. Unfortunately, we couldn't study the temperature dependence of the peak measured at (½, ½, ½) since we also measured a cuprate thin film to benchmark the technique. We hope that other beam time will be granted to this project, as it will enable us to address a significant question regarding superconductivity in this novel system.

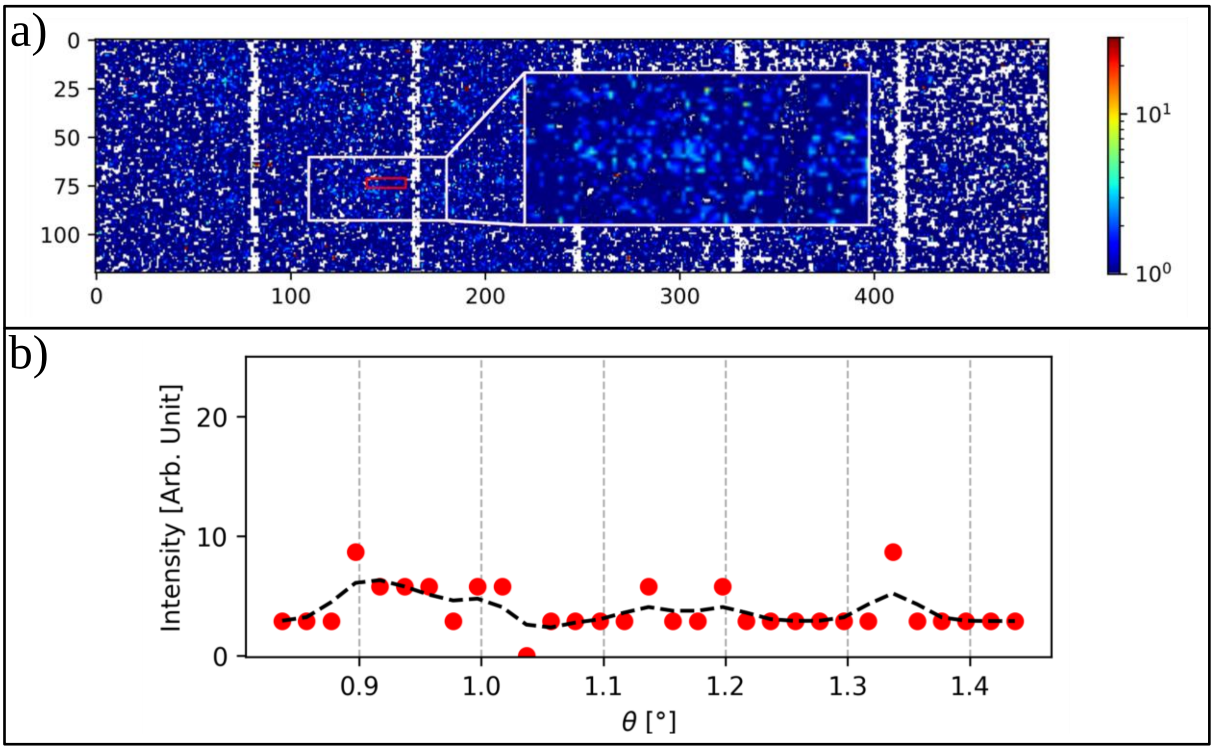

Figure 2: a) Detector image and b) transverse scan of scattered X-ray at 300K and (½, ½, ½) Bragg point. The experimental measurement parameter in this figure is the same as Figure 1 , with the exception of the temperature change.

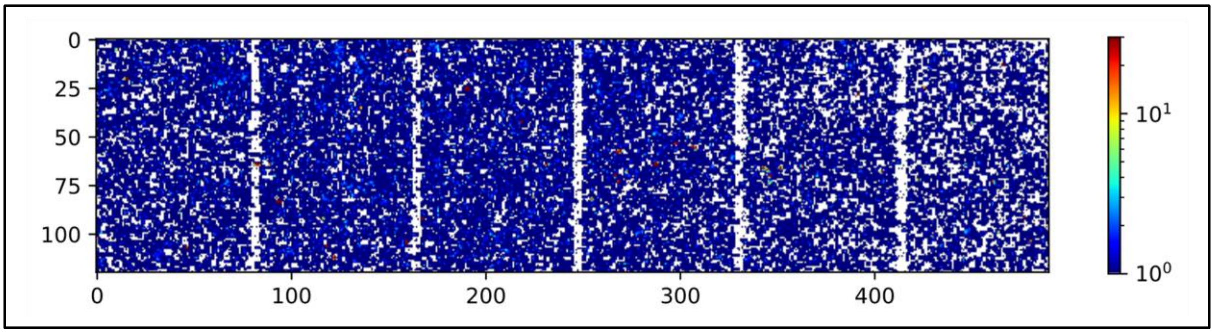

Figure 3: Detector image of scattered X-ray at 20K and (½, ½, 3/2) Bragg point.

- Justification and comments about the use of beam time (5 lines max.): **-**

During this beam time we had no interruption, and everything was working nicely. Also, we had a very nice support from the local contact that allowed as to easily analyse the data by using Python.

- Publication(s): **-**