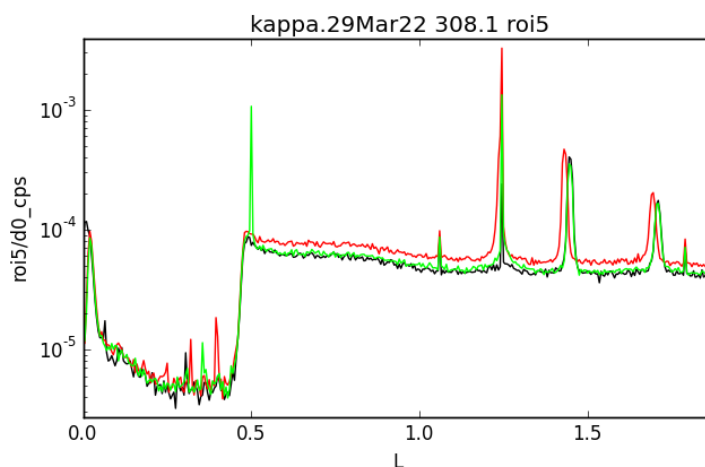


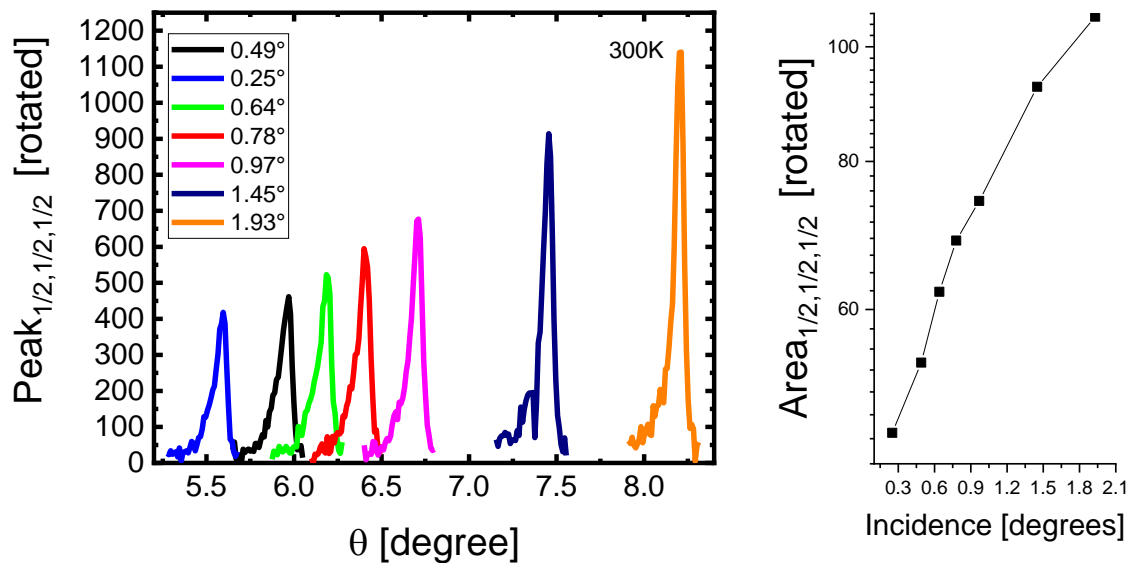
## Experiment report for Proposal A02-2-880

Recently, the discovery of superconductivity in infinite-layer nickelate thin films has generated a great excitement, as their properties are believed to be related to the  $\text{NiO}_2$  plane, which are considered equivalent to the  $\text{CuO}_2$  planes in cuprates. Recently it has been reported that infinite layer nickelates exhibit peculiar magnetic excitations with a bandwidth of circa 200 meV in  $\text{SrTiO}_3$ -capped  $\text{NdNiO}_2$  thin films. Here we are looking for the direct evidence of long-range magnetic order in infinite nickelate thin films, for which a high grazing geometry is then required. Although we could find in this unusual geometry a peak in the rotated channel, with almost no temperature dependence in the accessed range (10-300 K), we cannot firmly assume that this it is related to a peak stemming from the expected long-range magnetic order in infinite-layer nickelate thin films. The main concern of the Team that has conducted the experiment, is about the nature of the Crystal Truncation Rod (CTR) measurement which rendered around the  $(-0.5 \ 0.5 \ 0.5)$  position a practically one single pixel-like peak, please see figure below (green-curve).



Resonant Elastic X-ray Scattering (REXS) and Surface Resonant X-Ray Diffraction (SRXRD) experiments were performed on the D2AM beamline equipped with a cryofurnace. The sample was a 10-nm-thick films of  $\text{NdNiO}_2$  grown on a  $\text{SrTiO}_3$  (001) substrate with a  $\text{SrTiO}_3$  capping layer deposited by pulsed laser deposition and on which at ID32 in a previous RIXS experiment we could observe clear signatures of spin-excitation<sup>1</sup>. X-ray Resonant Magnetic Scattering (XRMS) measurements were carried out at the energy of the Ni K-edge around the  $(-1/2, 1/2, L)$  crystal truncation rod (CTR). First, we have tested that we could obtain signatures of a

magnetic diffraction peak for the prototypical NiO single crystal in a very high grating geometry, and we could get similar results as Barbier *et al.* Basically we measured the  $[\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$  antiferromagnetic diffraction plane in grazing scattering geometry at 300 K<sup>2</sup>. The results are shown in the figure below. Although less intense we could nevertheless observe the AFM peak in the grazing geometry, which confirmed that we can use this set-up geometry for our nickelate system that can be synthesized uniquely as a thin film. However, given the doubtful result, we thought that it would be better to benchmark the set-up for a thin film system where the long-range AFM is very well known.



Therefore, the energy scan was acquired at Ni K-edge in the energy range of 8.327 keV to 8.346 keV in a very grazing geometry for the infinite-layer sample. We can see in Fig.1 that the peak is resonating at the Ni K-edge even after mounting the polarization analyzer.

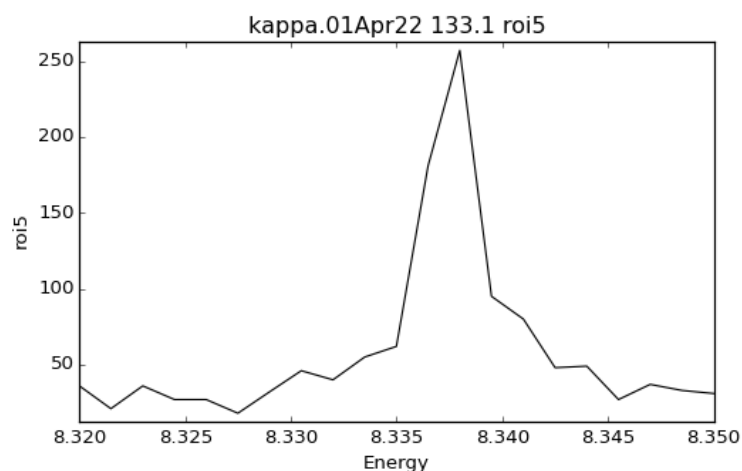


Fig.- 1 Energy scan around Ni K-edge in grazing geometry.

Figure 2 shows the feature observed at the  $[-\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$  peak position at 6K in the rotated channel.

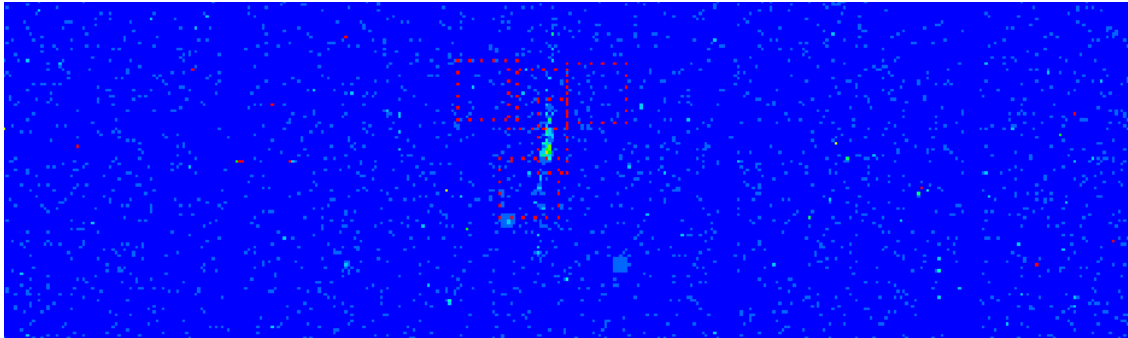


Fig.- 2  $(-\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  peak at 6 K in the rotated channel.

SRXRD spectra were recorded at seven different temperatures (6, 28.6, 70, 89, 128, 228, and 300 K). Fig.3 shows the temperature dependent area of the  $(-\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  peak with analyzer. We did not observe any decrease of the peak intensity even at higher temperature values, *i.e.* 600 K. Further measurements are required to make a final decision about our results.

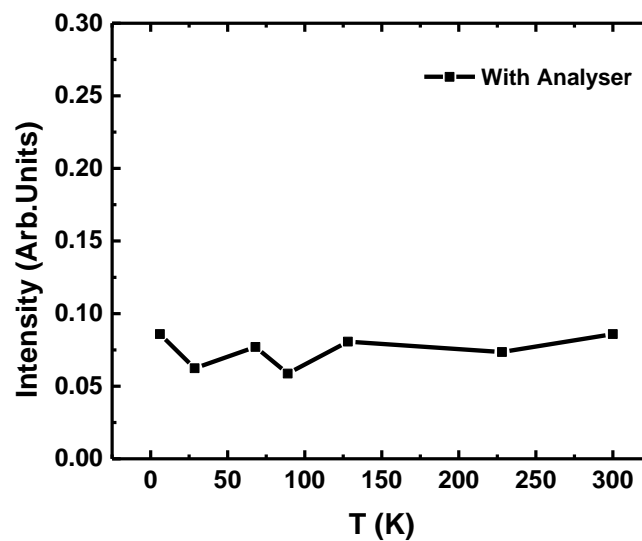


Figure 3 Temperature dependence of the  $(-\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  peak area.

1. Krieger, G. *et al.* Charge and Spin Order Dichotomy in NdNiO<sub>2</sub> Driven by the Capping Layer. *Phys. Rev. Lett.* **129**, 27002 (2022).
2. Barbier, A. *et al.* Surface and Bulk Spin Ordering of Antiferromagnetic Materials: NiO(111). *Phys. Rev. Lett.* **93**, 257208 (2004).