

## 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://www.esrf.fr/misapps/SMISWebClient/protected/welcome.do>

### ***Reports supporting requests for additional beam time***

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

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.

### **Deadlines for submission of Experimental Reports**

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

### **Instructions for preparing your Report**

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal 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.



	<b>Experiment title:</b> Magnetic excitations in RNiO <sub>3</sub> thin films and heterostructures studied by Ni-L <sub>3</sub> RIXS	<b>Experiment number:</b> HC3012
<b>Beamline:</b>	<b>Date of experiment:</b> from: 19/04/2017 to: 25/04/2017	<b>Date of report:</b> 6/6/2017
<b>Shifts:</b>	<b>Local contact(s):</b> BETTO Davide	<i>Received at ESRF:</i>
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## Report:

The aim of the current experiment was to measure for the first time the magnetic excitations in RNiO<sub>3</sub> thin films and heterostructures by exploiting the high resolution and sensitivity of the new ERIXS spectrometer at the ID32 beamline.

The antiferromagnetic (AF) phase of bulk nickelates is characterized by an unusual  $q_{AF}=(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})_{pc}$  ordering vector and a non-collinear spin spiral, which can be tuned by epitaxial strain and spatial confinement. Despite the extensive research that has been performed on RNiO<sub>3</sub> so far a direct experimental characterization of the magnetic excitations is still missing [1,2].

For this reason, we prepared high-quality thin films of the well-studied nickelates NdNiO<sub>3</sub> (NNO), PrNiO<sub>3</sub> (PNO) and LaNiO<sub>3</sub> (LNO). In addition to the bulk samples, we also brought superlattices with different periodicities comprising RNiO<sub>3</sub> as active layer and RAlO<sub>3</sub> as insulating buffer layers. Thanks to the unprecedented resolving power of the ERIXS spectrometer, we report here the first successful observation of low energy (<100meV) dispersive magnetic excitations in 113 nickelates (see Fig. 1).

During our experiment the sample was first aligned to the maximum of the characteristic AF Bragg peak  $q_{AF}=(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})_{pc}$ . The incident energy was tuned to the Ni  $L_3$  -edge to maximize sensitivity and intensity of the magnetic signal. We note here, that due to the quite low counting rate at the Ni  $L_3$  -edge each spectrum had to be collected over three hours. The combined resolution was  $\sim 50\text{meV}$ , *i.e.* sufficient to detect the low energy magnons in RNiO<sub>3</sub> while allowing for reasonable accumulation time. Among others, we measured the magnetic dispersion along the  $(111)_{pc}$  direction (that is the direction of the AF wavevector) in the accessible  $q$ -range from  $q=0.4 \text{ \AA}^{-1}$  (far away from  $q_{AF}$ : blue in Figs. 1 and 2) to  $q=0.65 \text{ \AA}^{-1}$  (close to  $q_{AF}$ : red in Figs. 1 and 2). As illustrated in Fig. 1, it is necessary to measure the

dispersion both well below and above the AF transition temperature in order to decompose the low energy part into elastic, phononic and magnetic contribution.

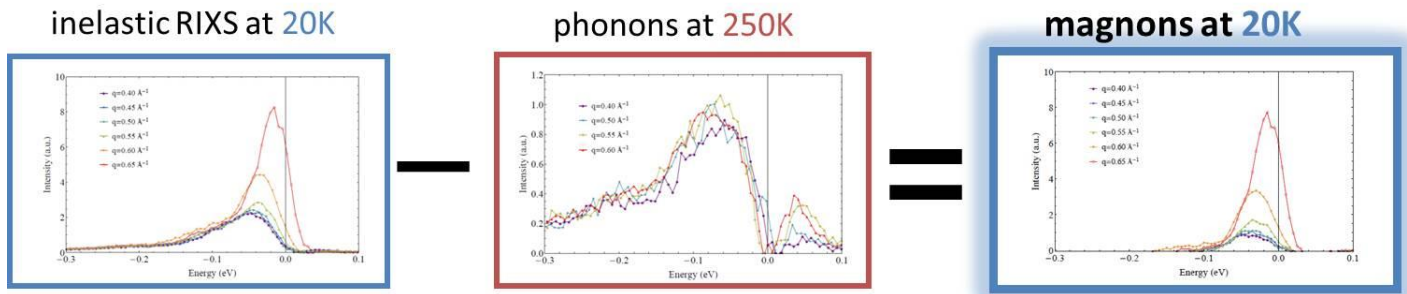


Figure 1: Illustration of spectral decomposition and magnetic dispersion for the NNO thin film.

We collected systematic datasets for several samples. Figs. 1 and 2 show representative magnetic dispersions for bulk NNO and for a PNO/PAO superlattice, respectively. For all samples, we observed a magnon with an energy dispersion of approx. 30meV and an increased spectral weight close to  $q_{AF}$ , which make sense with the expectations for magnetism in  $RNiO_3$ . Our results will allow for the first time the extraction of exchange coupling parameters which remained to date unknown by combining the experimental findings with a theoretical model currently under development in our group. Our preliminary spin-wave calculations based on a Heisenberg-like Hamiltonian reveals coupling constants of a few meV for the NNO film. The possibility to study the magnetic exchange interactions in 113-nickelates and compare them to cuprates will thus provide new insights into the properties of these materials and on the possibility of achieving artificial superconducting nickelate heterostructures.

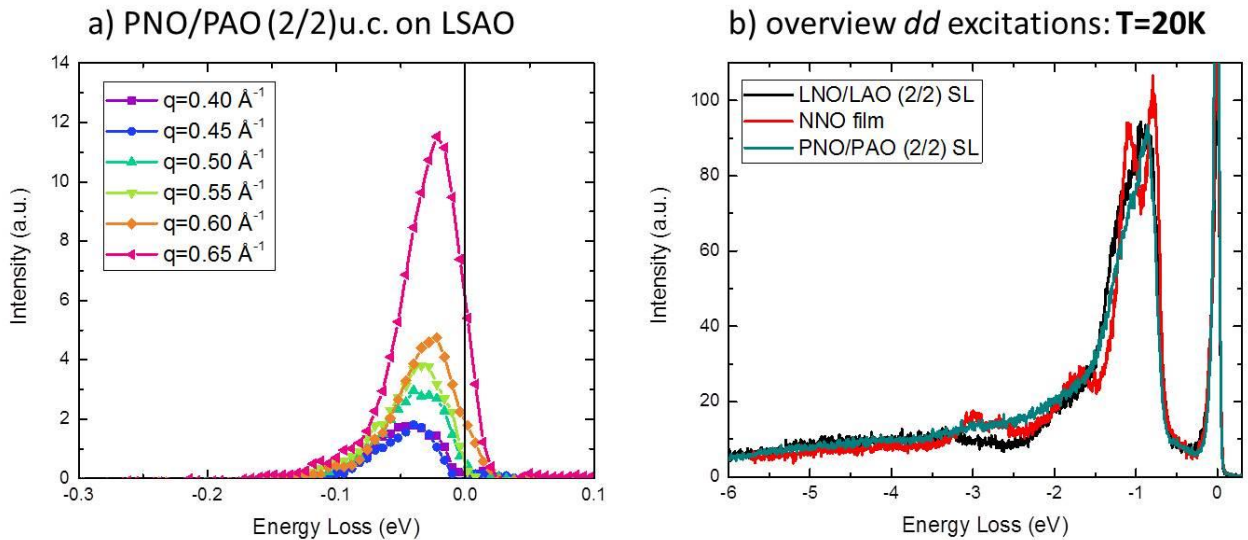


Figure 2: a) Magnetic dispersions at  $T=20K$  observed in PNO/PAO-based superlattices. b) Overview of RIXS spectra for different  $RNiO_3$ -based thin films and heterostructures.

We also measured RIXS spectra of LNO as a function of dimensionality: in bulk form and as part of a LNO/LAO superlattice [3]. In fact we were able to collect some preliminary spectra on a single crystal, which was successfully grown only very recently [4]. However, the analysis of the low energy part of spectrum of LNO is complicated by the presence of a

strong continuum most probably due to charge excitations and to the strong metallicity of LNO, which is presumably damping the magnetic excitations.

In addition to the low energy magnons discussed above, we got simultaneously systematic data on the high energy charge (*dd*) excitations for the three prototypical  $R\text{NiO}_3$ ,  $R=\text{Pr}$ ,  $\text{Nd}$  and  $\text{La}$  (see Figure 2b). As the experiments were carried out only very recently, our analysis of the *dd* excitations remains to be finalized: we can anticipate that we were able to observe pronounced changes in the high energy spectrum as a function of rare-earth radius (and consequently Ni-O bonding angle), spatial confinement and epitaxial strain.

To conclude, we report the first experimental observation of magnetic excitations in 113 nickelates. Our results are a fundamental step to understand the magnetic dynamics in  $R\text{NiO}_3$  and facilitate for the first time a sound theoretical modelling. Following the results shown above, we are currently preparing a manuscript for publication.

[1] Fano *et al.*, Phys. Rev. Lett. **111**, 106804 (2013)

[2] Hepting *et al.*, Phys. Rev. Lett. **113**, 227206 (2014)

[3] Boris *et al.*, Science **332**, 6032 (2011)

[4] Li *et al.*, arXiv 1705.0289

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other suggestion for Fig. 1

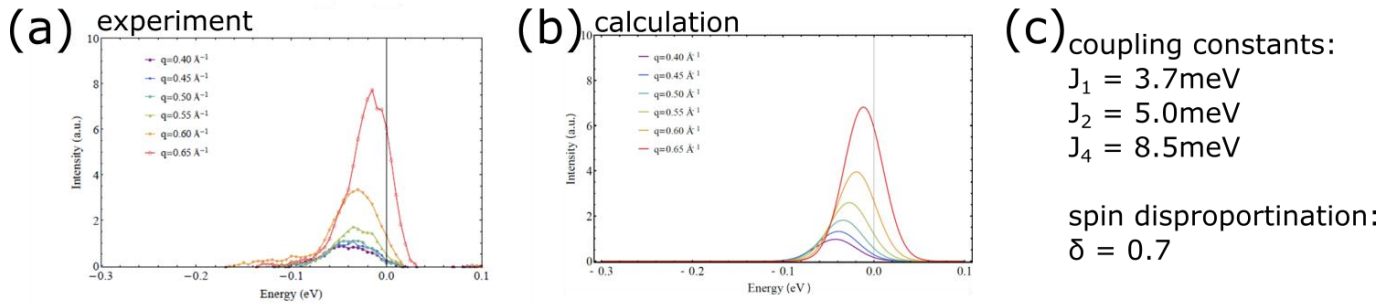


Figure 1: Spin excitations probed by Ni  $L_3$  -edge RIXS for a NNO thin film a) Experimental magnetic dispersion. We extracted the magnetic dispersion from the quasi-elastic RIXS signal by eliminating the elastic and phononic contribution. b) Calculated magnetic dispersion. The calculated magnetic dispersion with the parameters given in c matches nicely to the experimental one. c) Extracted model parameters. The preliminary coupling constants are extracted by fitting the theoretical dispersion to the experimental. For the first time, the magnetic exchange interaction in  $R\text{NiO}_3$  was quantified facilitating a directly comparison to the coupling constants of cuprates.

other suggestion for Fig. 2

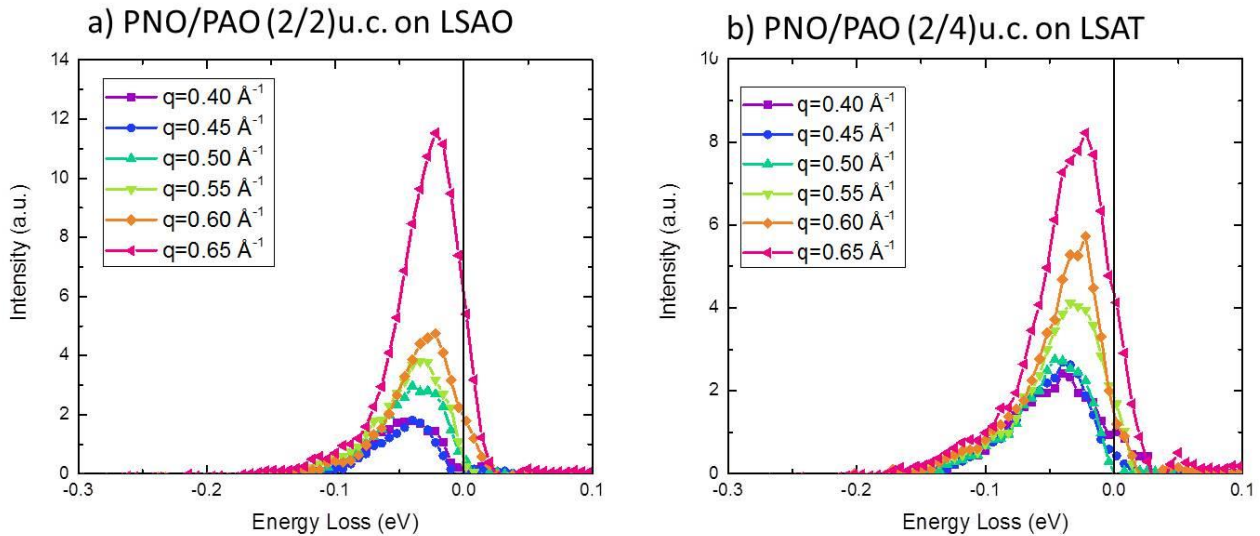


Figure 2: Magnetic dispersions at  $T=20\text{K}$  observed in PNO/PAO-based superlattices. At this early stage of our analysis we resort to the graphs shown above and do not yet discuss detailed effects of epitaxial strain and 2D confinement on the magnetization dynamics.