EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

ESRF

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:** Spin-orbital excitations in the j=1/2 Mott insulators α-Li_2IrO_3 and β-Li_2IrO_3	**Experiment number**: HC-3764
Beamline: ID20	**Date of experiment**: from: 11/04/2018 to: 17/04/2018	**Date of report**:
Shifts: 18	**Local contact(s)**: Marco Moretti	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

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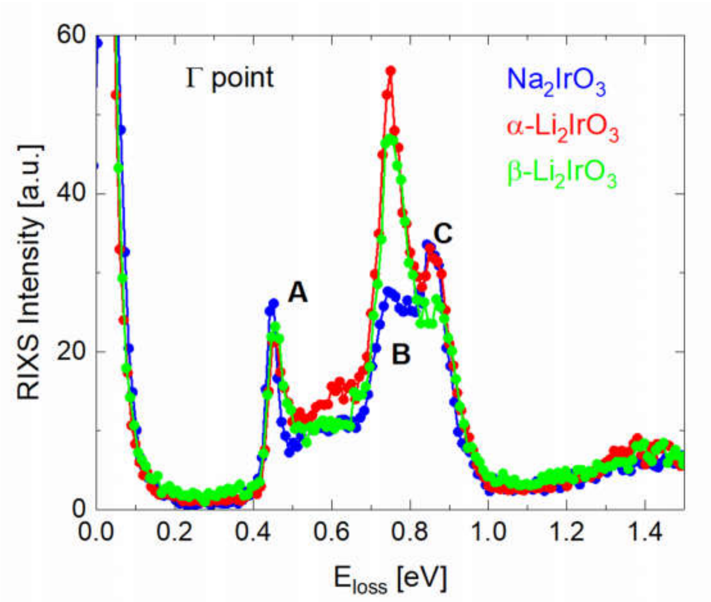
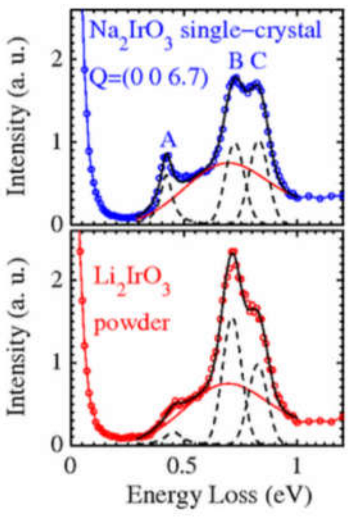
Report:

High-resolution RIXS has been measured at the Ir L_3 edge at T = 10 K on the tricoordinated Mott-insulating iridates α-Li_2IrO_3, Na_2IrO_3, and β-Li_2IrO_3. The first two show a monoclinic lattice structure featuring stacks of quasi-2D honeycomb layers. Samples of both compounds show stacking faults that lead to the formation of three different domains. By scanning the samples' surface we found a spot where a single domain was dominant (> 90%). In the case of Na_2IrO_3, we measured on an *ab* surface with *b* in the scattering plane. RIXS data of α-Li_2IrO_3 were measured on an *ab* surface with the *b* axis forming an angle of 60° with the scattering plane. The compound β-Li_2IrO_3 shows an orthorhombic crystal structure. Here, we measured on an *ab* surface with *b* perpendicular to the scattering plane.





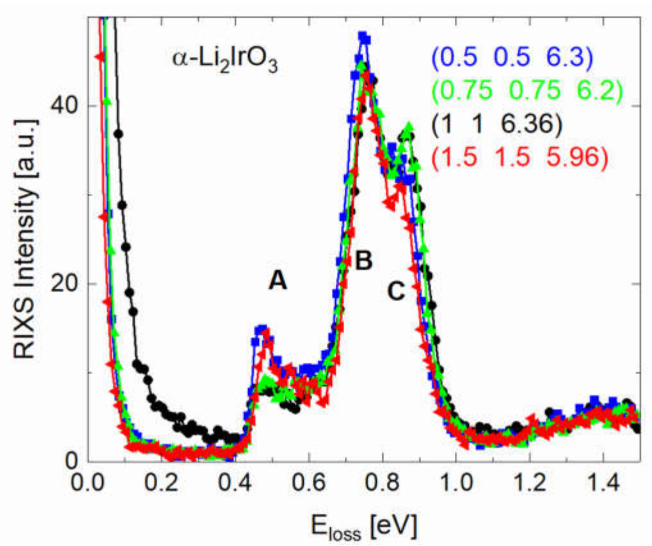
Gretarsson *et al.* [1]

Previous RIXS data on single crystals of Na_2IrO_3 by Gretarsson *et al.* [1] revealed three peaks denoted as **A**, **B**, and **C**, see figure on bottom right on p. 1. Peaks **B** and **C** were attributed [1] to the spin-orbit exciton, i.e., the excitation from the j=1/2 ground state to the j=3/2 excited state. The peak splitting signals a non-cubic crystal field. Furthermore, Gretarsson *et al.* [1] interpreted peak **A** as an exciton in the vicinity of the Mott gap. RIXS data on polycrystalline α-Li_2IrO_3 show similar features but a much broader peak **A** [1].

Our RIXS data on Na_2IrO_3 are consistent with those measured by Gretarsson *et al.* [1]. Remarkably, our data on single crystals of α-Li_2IrO_3 and β-Li_2IrO_3 reveal a stunning similarity between all three compounds, see figure on bottom left of p. 1. The energies of peaks **A**, **B**, and **C** are very similar in all three compounds, In contrast to the powder data of Gretarsson *et al.* [1], the single-crystal data on α-Li_2IrO_3 show a very sharp peak **A**, in close agreement with the other compounds.

The spin-orbit exciton and thus peaks **B** and **C** can be reasonably well described by a simple picture of individual Ir ions in a non-cubic crystal field. The observation that peaks **B** and **C** are located at the same energy for all three compounds immediately tells us that both the non-cubic crystal field and the size of spin-orbit coupling are very similar in all three compounds, despite the different crystal structures.

The excitonic interpretation of peak **A** requires to go beyond a simple single-site picture, In agreement with the data of Gretarsson *et al.* [1] on Na_2IrO_3, the spectra we measured on α-Li_2IrO_3 at different transferred momentum **q** show negligible dispersion of all three excitations, see figure on p. 2. For peaks **B** and **C**, this agrees with the predominantly local character of the spin-orbit exciton. However, it also indicates a local character of peak **A**. Furthermore, we observe that the intensity of peak **A** shows a strong dependence on the transferred momentum **q**, see figure. This explains why peak **A** was less pronounced in the powder data, averaging over all directions of **q**. It furthermore will allow us to reveal the precise character of peak **A** based on a detailed analysis of the observed **q** dependence. We will also compare the energy of peak **A** determined in the RIXS measurement with results of optical measurements for the Mott gap.



[1] Gretarsson *et al.*, Phys. Rev. Lett. **110**, 076402 (2013)