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:** RIXS on fcc Heisenberg-Kitaev materials with potentially ideal j=1/2 moments	**Experiment number**: HC-4491
Beamline: ID20	**Date of experiment**: from: 07.04.2021 to: 12.04.2021	**Date of report**: 4.3.2023
Shifts: 15	**Local contact(s)**: Christoph Sahle	*Received at ESRF:*

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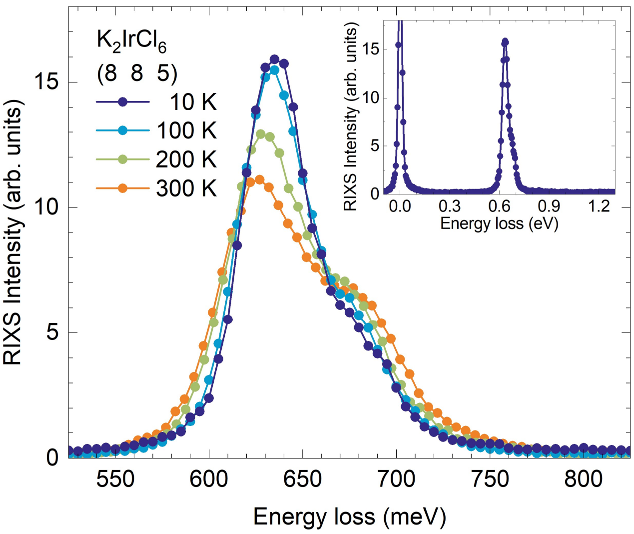
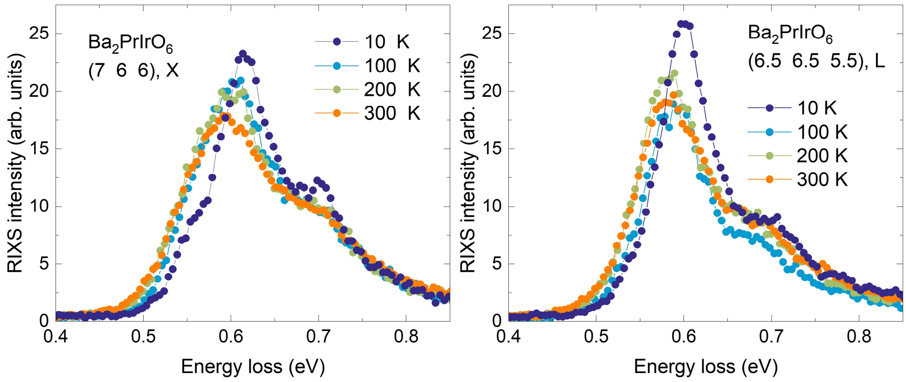
Report:

The present study is motivated by the search for an ideal realization of j = ½ moments in $5d^5$ iridates. These spin-orbit-entangled moments are the microscopic source of, e.g., Kitaev exchange on a tricoordinated lattice. The ideal j=½ wave function is found in a *cubic* crystal field while non-cubic contributions yield a finite admixture of j = 3/2 character. RIXS at the Ir L_3 edge is a powerful tool to detect such deviations from cubic symmetry via the related splitting of the spin-orbit exciton, i.e., the excitation from the local j = ½ ground state to a j = 3/2 excited state. Cubic symmetry on the Ir site is found in structural studies of, e.g., Ba_2CeIrO_6 where RIXS nevertheless shows a splitting of the spin-orbit exciton of about 100 meV [1]. The discrepancy can be explained by assuming that the distortions are local, exhibiting a negligible correlation length, such that the average global structure seen in elastic scattering still shows cubic symmetry. This, however, raises a fundamental question: Is there an electronic mechanism that drives j = ½ systems away from cubic symmetry? For Ba_2CeIrO_6, one example could be magneto-elastic coupling, lifting the frustration on the *fcc* lattice [1]. Even small deviations from the j = ½ wave function – with a small cost of lattice energy – may give rise to massive bond-dependent changes of exchange couplings and a large gain in magnetic energy. In the quest for exotic j = ½ spin liquids it is of general importance to understand whether j = ½ compounds avoid frustration in this way. An alternative explanation may be the presence of defects. To adress this issue, we measured RIXS at the L_3 edge on two globally cubic (*fcc*) $5d^5$ model systems for frustrated quantum magnetism with potentially ideal j = ½ moments, antifluorite-type K_2IrCl_6 and the double perovskite Ba_2PrIrO_6. In both cases, we measured on a (111) surface with an energy resolution of about 25 meV.



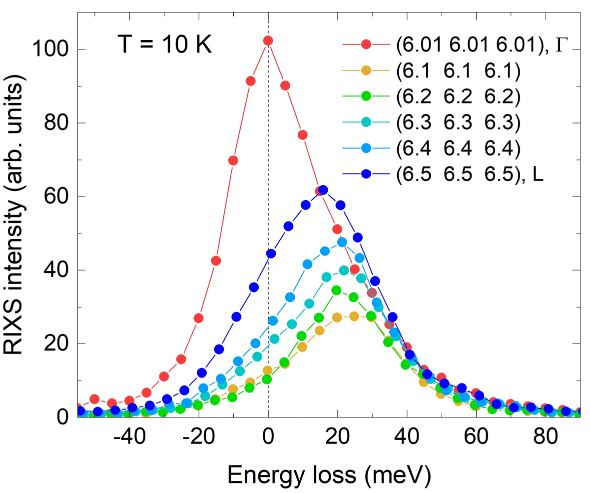
In K_2IrCl_6, we focus on the temperature dependence since the spitting of the (rather local) spin-orbit exciton should not depend strongly on **q**, in agreement with the results on Ba_2CeIrO_6 [1]. Moreover, the Mott gap of K_2IrCl_6 is larger than 1 eV, which allowed us to study the spin-orbit excition also in infrared transmittance, where we performed a thorough analysis of the temperature dependence. The infrared data show strong phonon sidebands, and the spin-orbit exciton can only be studied in a phonon-assisted process, offering complementary information to RIXS. For a meaningful comparison of the two data sets, we measured RIXS at 10, 100, 200, and 300 K at the X point (8 8 5), see Fig. 1 above. At 10 K, the RIXS data are dominated by a single peak at 0.64 eV. However, also K_2IrCl_6 shows an additional (weak) feature at about 0.68 eV, and the relative intensity of this mode rises with increasing temperature. We expect that a combined analysis of RIXS and optics will yield the strength of the coupling to phonons and unravel the role of magneto-elastic coupling.

The double perovskite Ba_2PrIrO_6 orders magnetically at T_N = 71 K. Note that this implies a reduction of the crystal symmetry below T_N. Here, we also measured RIXS data at 10, 100, 200, and 300 K, focusing on the X point (7 6 6) and the L point (6.5 6.5 5.5), see Figure 2 below. The measurements were time consuming since we used a step size of 5 meV and had to average over several scans to obtain a smooth data set with well-defined features. Like K_2IrCl_6, also Ba_2PrIrO_6 shows a splitting of the spin-orbit exciton, a remarkable result. At the L point, we observe one dominant peak with only a small second mode, similar to the data of K_2IrCl_6 (there at the X point). At the X point, however, Ba_2PrIrO_6 shows a further feature on the low-energy side which is most pronounced at 10 K. It is tempting to speculate that such additional features are caused by the interaction with the Pr j = 5/2 moments. Furthermore, the splitting of the spin-orbit exciton in the oxides Ba_2PrIrO_6 and Ba_2CeIrO_6 equals about 100 meV and is thus roughly a factor 2 larger than in the chloride K_2IrCl_6, pointing towards a possible origin in terms of phonon sidebands. A thorough analysis will have to show whether the lineshape can be related to the (highest) phonon energies in these compounds.



Remarkably, both globally cubic compounds studied here exhibit a splitting of the spin-orbit exciton. We expect that these results, in combination with our optical data, will allow us to unravel the origin of this surprising behavior.

Using the spin-orbit exciton in Ba_2PrIrO_6 as an example for a rather local excitation, we studied the possibility to measure with four spherically diced analyzer crystals in parallel. In principle, each analyzer yields a signal on a separate part of the 2D detector. The aim was to enhance the efficiency of the beam time, acquiring data at different **q** in parallel. However, we found that spurious signals from the additional analyzers spill into the signal, giving rise to artifacts such as small "sidebands" in the line shape. Since the present study is focussed on the line shape of the spin-orbit excition, we continued the measurements with a single analyzer.



A further goal was to find signatures of magnons in Ba_2PrIrO_6 since the high ordering temperature 71 K points towards large exchange couplings. The figure shows low-energy data from Γ to L at 10 K. The elastic line is suppressed for $2\theta = 90°$. The data at (6.2 6.2 6.2) are closest to this ideal case and indeed show the smallest intensity at zero loss. Note that the zero of energy loss was calibrated by reference measurements with a large elastic line. We find a magnon peak at about 20 meV that shows a small dispersion. Close to Γ, at (6.01 6.01 6.01), the elastic line dominates and corroborates the position of zero loss. Resolving these low-energy magnons in Ir *L* edge RIXS is a major success of this beam time.

[1] A. Revelli *et al.*, Phys. Rev. B **100**, 085139 (2019).