ESRF	**Experiment title:** Spin-orbit excitons in j=1/2 triangular lattice iridates	**Experiment number**: HC-2880
Beamline: ID20	**Date of experiment**: from: 25.1.2017 to: 31.1.2017	**Date of report**: 24.2.2017
Shifts: 18	**Local contact(s)**: Marco Moretti	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Paul van Loosdrecht * II. Physikalisches Institut, University of Cologne

Markus Grüninger * II. Physikalisches Institut, University of Cologne

Marco Moretti * ESRF Grenoble **Jeroen van den Brink** IFW Dresden

Giulio Monaco * Dipartimento di Fisica , Universita di Trento

Raphael German * II. Physikalisches Institut, University of Cologne

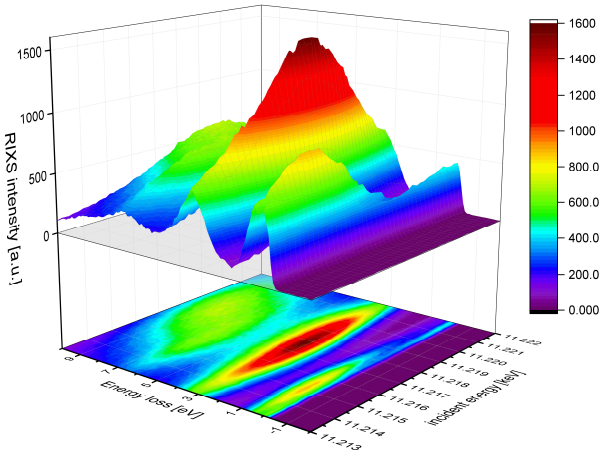
Thomas Koethe * II. Physikalisches Institut, University of Cologne

Krzysztof Wohlfeld University of Warsaw Faculty of Physics

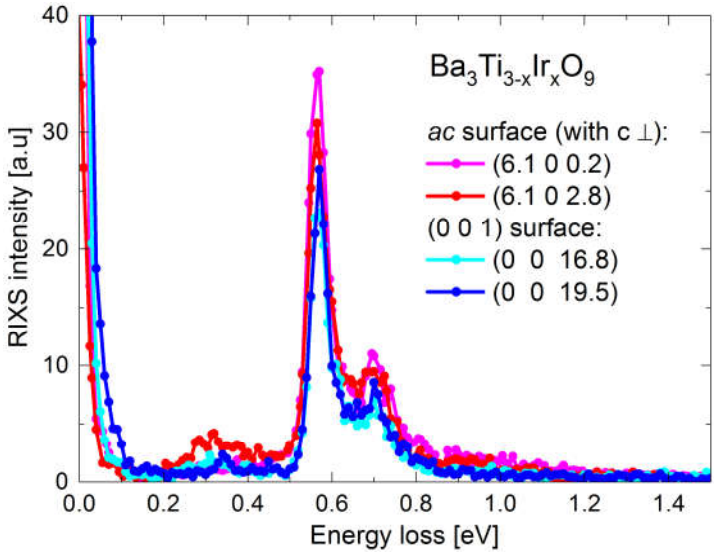
Report:

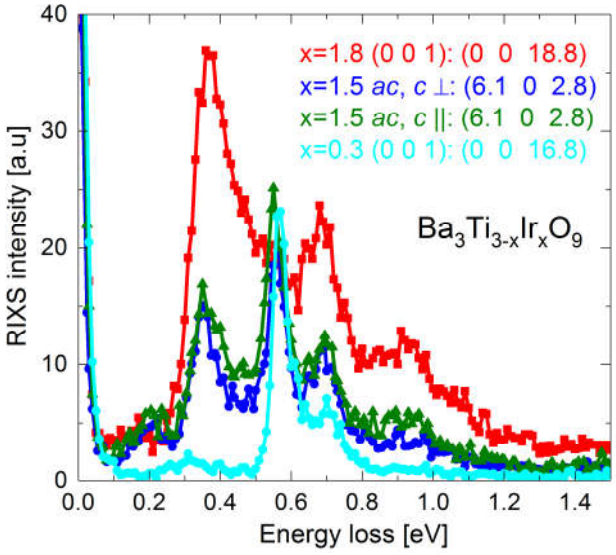
High-resolution RIXS has been measured at the Ir L_3 edge on different specimen of the triangular-lattice iridate family $Ba_3M_{3-x}Ir_xO_9$ (hexagonal space group $P6_3/mmc$). We focussed on M = Ti^{4+} (different x as determined by XRD and/or EDX as well as different orientations), performed some additional measurements for M = Ce (x=2) on a (0 0 1) surface, and collected a few exploratory scans on M = Sc (x=1.5, with $5d^4$ Ir^{5+} ions). These compounds consist of triangular layers of $(M,Ir)_2O_9$ bioctahedra in which the two octahedra are connected in a face-sharing way. The $5d^5$ iridates are candidate materials for novel quantum states of matter based on exotic bond-directional exchange interactions in spin-orbit entangled j=1/2 Mott insulators. For the realization of such novel quantum states, it is important to understand how e.g. crystal field and hopping interactions affect the j=1/2 character. RIXS at the Ir L edge is the ideal tool to determine the j=1/2 character by measuring the low-energy excitations.

To determine the resonance behavior, we collected a low-resolution (0.4 eV) RIXS map at room temperature on $Ba_3Ti_{1.2}Ir_{1.8}O_9$, see figure below. The data show intra-t_{2g} excitations below 1 eV, excitations into e_g levels above about 3 eV, and charge-transfer excitations at still higher energies. In the following, high-resolution data (27 meV) were collected at T=20K with an incident energy of 11.215 keV for M = Ti (11.2155 keV for M = Ce or Sc) in order to resonantly enhance the t_{2g} excitations.



For x = 0.3 and 0.5 and M = Ti, RIXS measurements show the expected response of the excitations from the j=1/2 ground-state doublet to the excited j=3/2 quartet which however is split by the trigonal crystal field. The splitting equals about 130 meV. The RIXS data are very similar for





measurements on an (0 0 1) surface and an *ac* surface and hardly vary as a function of the transferred momentum. Exemplary data are shown in the top left figure. This result provides a textbook example of the spin-orbit exciton, showing a very narrow line width. The data will allow us to determine the spin-orbit coupling parameter λ as well as the strength of the trigonal crystal field.

For larger Ir concentration *x*, the probability to find Ir_2O_9 bioctahedra (instead of $TiIrO_9$) increases and the excitation spectrum changes dramatically, see top right figure. This reflects the strong change of the ground state due to hopping between the two Ir sites within the face-sharing octahedra, suggesting the formation of molecular orbitals. Qualitatively, the data show a smooth crossover from diluted j=1/2 moments at small *x* to molecular orbitals at large *x*. We will perform quantitative theoretical calculations to determine the relevant parameters as well as the character of the ground state. The formation of molecular orbitals with S=0 on the one hand and the observation that local j=1/2 moments dominate only in the dilute limit on the other hand are important to understand the small magnetic susceptibility of these compounds and the concomitant claims of spin-liquid behavior. Additionally, we find that the spectral weight of the low-energy excitations depends sensitively on the transferred momentum, see bottom left figure.

In the case of $Ba_3CeIr_2O_9$, the RIXS features are shifted to higher energies compared to *M* = Ti, see bottom right figure. The line shape can be interpreted in terms of molecular orbitals, and the shift in energy can be attributed to the enhanced hopping for *M* = Ce. Similar to *M* = Ti, we observe a pronounced change of the RIXS intensity as a function of the transferred momentum. In $Ba_3Sc_{1.5}Ir_{1.5}O_9$ with Ir^{5+} ions we finally observe a series of broad RIXS features at intermediate energies compared to the results for *M* = Ti and Ce.

Currently, we perform a detailed quantitative analysis of the measured data. These results clarify the local electronic structure of $Ba_3M_{3-x}Ir_xO_9$ and will contribute to a more thorough understanding of j=1/2 iridates. Due to the richness of the physics and the large amount of high-quality data, we plan to publish the results in two separate publications, one on *M* = Ti and a second one on *M* = Ce and Sc.