ESRF	**Experiment title:** Elementary excitations in spin-orbit induced Mott insulator $CaIrO_3$ by resonant inelastic hard x-ray scattering	**Experiment number**: HC738
Beamline: ID20	**Date of experiment**: from: 17/07/2011 to: 23/07/2011	**Date of report**: 19/09/2013
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Report:

Contrary to the naive expectation that 5d transition metal oxide exhibits metallic behavior owing to the weak correlation effect, a postperovskite-type iridium oxide $CaIrO_3$ is an antiferromagnetic Mott insulator [1,2]. The underlying mechanism of this insulating state was considered to be the band reconstruction by the strong spin-orbit coupling in 5d transition metals: the Ir t_{2g} orbitals are reconstructed by the spin-orbit coupling into the two or three orbitals with narrower bandwidth, which drives the transition from a correlated metal into a Mott insulator [3-6]. To check this mechanism, we need to fully reveal the energy scheme of t_{2g} orbitals in $CaIrO_3$.

On the other hand, the superexchange interaction in iridium oxides is theoretically shown to be unique [7]. Whereas an antiferromagnetic Heisenberg interaction $J_1 S_i \cdot S_j$ is dominant in a corner-shared IrO_6 bond, the magnetic interaction across the edge-shared bond becomes a highly anisotropic and ferromagnetic one, $-J_2 S_{iz} S_{jz}$ (quantum compass model). Experimentally, these two types of superexchange interactions are realized in $CaIrO_3$, in which spins are aligned ferromagnetically (antiferromagnetically) along the edge- (corner-) sharing direction below 115 K. To gain further insights on these exchange interactions, it is highly expected to evaluate J_1 and J_2 values from the dispersion relation of magnons.

Motivated by these issues, we studied the electronic and magnetic elementary excitations of $CaIrO_3$ by means of the high resolution RIXS at the Ir L_3 edge; this powerful technique was applied to various iridium oxides to probe electronic and magnetic excitations [8-10]. In our experiments on ID20, the incident x-rays were monochromatised with a Si(111) double crystal premonochromator and with a Si(844) back-scattering channel-cut to a final bandwidth of 15 meV at 11.215 keV, which is the Ir L_3 edge energy. The beam was focused at the sample position down to a spot size of 10×20 μm^2. The spectrometer was based on the diced Si(844) analyser crystals (bending radius R = 2 m) and a pixelated position-sensitive detector to achieve a final energy resolution of ΔE ~ 25 meV. From a technical point of view, all the experiments were extremely successful; we have clearly observed electronic of d-d excitations as well as magnons at 30 K.

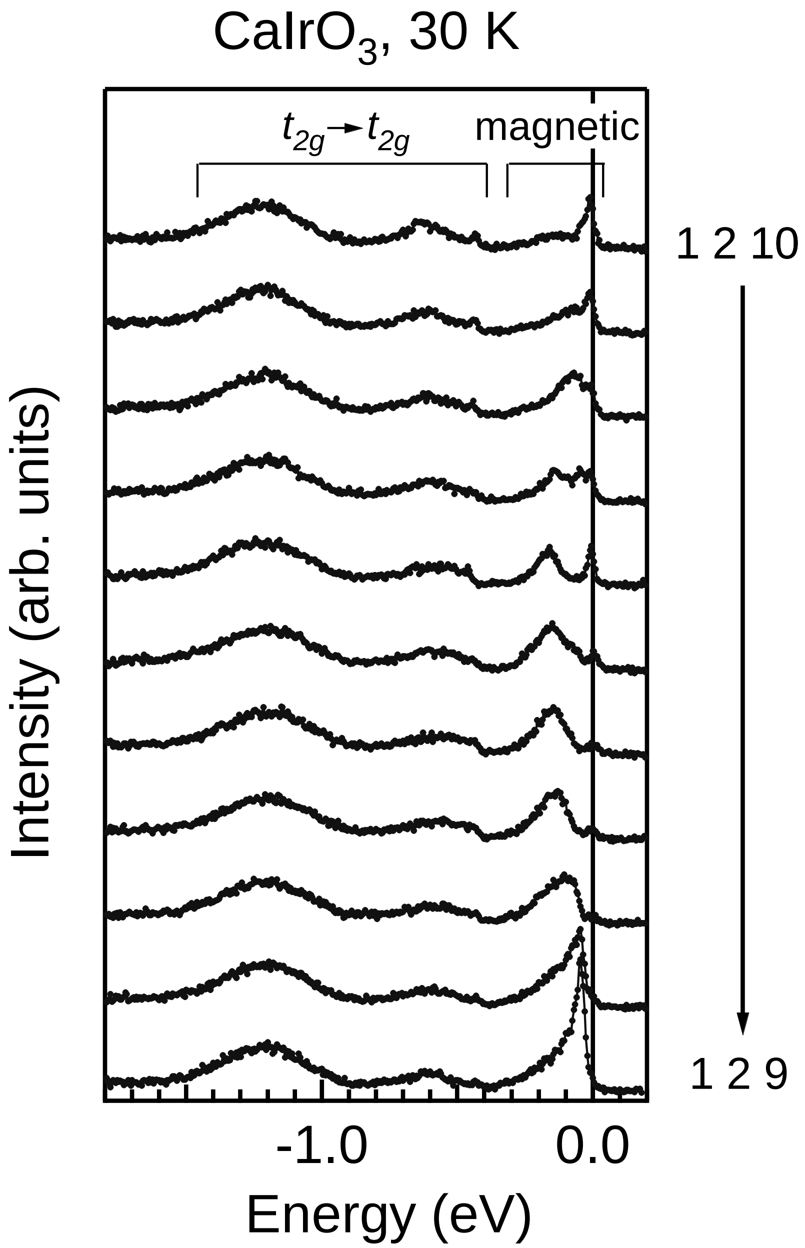

Fig. 1: Ir L3 edge RIXS spectra of CaIrO3 as a function of transferred momentum

Figure 1 is the typical example of Ir L_3 edge RIXS spectra for $CaIrO_3$. One can clearly see that there are two bands with small dispersions at 0.4–1.5 eV and a distinct band with sizable dispersions at 0–0.3 eV. The former ones are assigned to be the d-d excitations among t_{2g} orbitals, and the latter one is the magnon mode. Concerning the magnons, the measured dispersion curves as well as the scattering intensities are summarized in Fig.2 (the dashed line represents the energy resolution of the experiment).

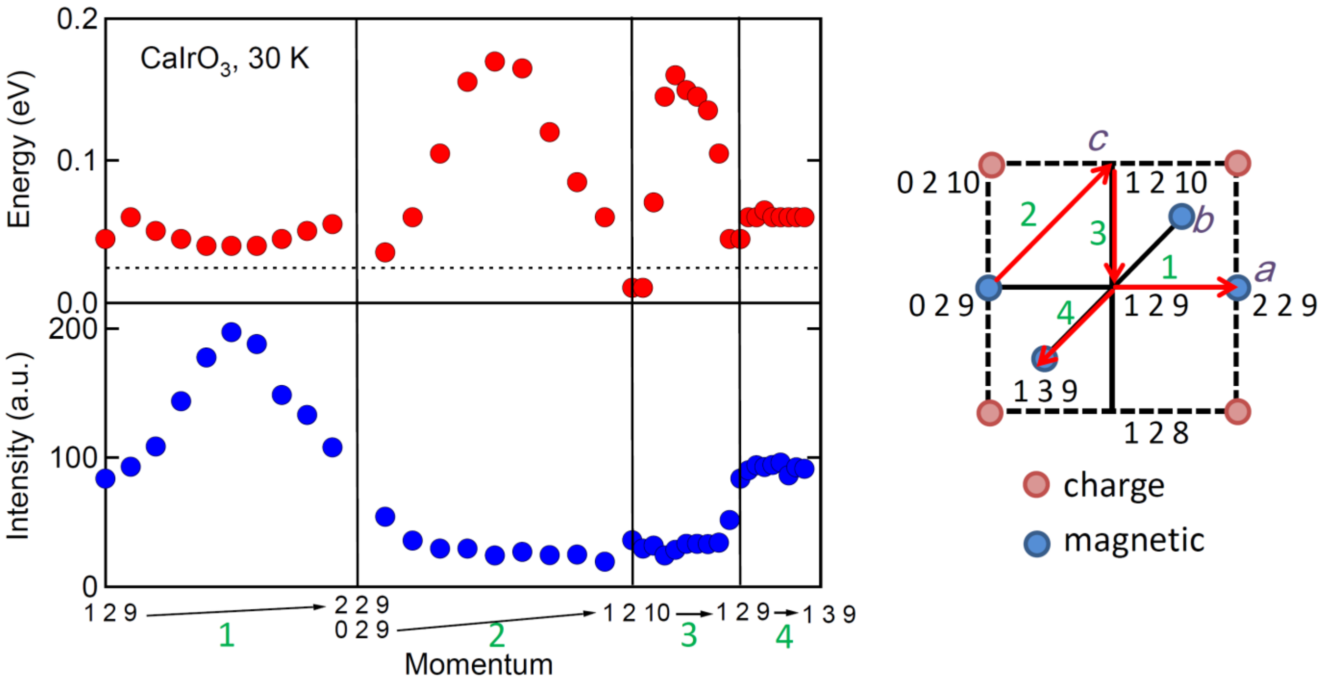

Fig. 2: Dispersion of magnetic excitations as a function of transferred momentum along high-symmetry directions in the Brillouin zone.

The striking features of our results can be summarized as follows:

- ✓ The d-d excitations associated with t_{2g} orbitals are split into two bands centered at 0.6 and 1.2 eV, indicating that the local tetragonal crystal field is comparable with the spin-orbit coupling.
- ✓ In the lowest energy regions of d-d excitations, there are excitonic features.
- ✓ The magnon modes show a small dispersion along the edge-sharing directions (a-axis), indicating the anisotropic quantum compass-type interaction.
- ✓ The magnon modes show a large dispersion along the corner-sharing directions (c-axis), indicating the isotropic Heisenberg-type interaction.
- ✓ The magnon modes show little dispersion along the layer direction (b-axis), indicating the weak interlayer coupling.
- ✓ The magnon modes in the lower energy region has larger intensities and longer lifetime in comparison with magnon modes in the higher energy region.
- ✓ Our preliminary analysis on the basis of the simple spin wave theory indicates J_1 ~140 meV and J_2 ~ 7 meV; $J_1 >> J_2$ is consistent with the proposed theory, in which J_2-term is produced by the perturbation of the Hund's coupling [7].

We are now working on a detailed interpretation of the experimental findings by collaborating with theoretical groups.

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