Experimental Report

09/07/2015

Proposal:	**4-01-1380**	**Council:**	4/2014

Title: Magnetic and Crystal Field Excitation Spectra in the Iridate Pyrochlores $Tb_2Ir_2O_7$ and $Er_2Ir_2O_7$

This proposal is a new proposal

Researh Area: Physics

Main proposer: **LEFRANCOIS Emilie**

Experimental Team:
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LHOTEL Elsa

Local Contact: OLLIVIER Jacques

Samples:
$Y_2Ir_2O_7$
$Tb_2Ir_2O_7$
$Er_2Ir_2O_7$

Instrument	Req. Days	All. Days	From	To
IN6	0	3	29/08/2014	01/09/2014
IN4	3	3	14/11/2014	17/11/2014

Abstract:

Our recent investigations of the magnetostatic properties of $Tb_2Ir_2O_7$ and $Er_2Ir_2O_7$ put into evidence different magnetic behaviours of Tb and Er in these iridate pyrochlores. The Ir-5d electrons exhibit a metal-insulator transition accompanied with a magnetic freezing at 130 K in $Tb_2Ir_2O_7$ and 140 K in $Er_2Ir_2O_7$. This magnetic freezing induces an "all-in all-out" magnetic order of the Tb moments in $Tb_2Ir_2O_7$ through the f-d exchange. In contrast, no Er magnetic order is detected by neutron diffraction in $Er_2Ir_2O_7$ down to 2K. We understand this by modelling the Tb magnetism with multi-axial Ising spins (aligned along the <111> directions) and the Er magnetism with multi-planar XY spins (perpendicular to these directions). As to confirm and further our insights, we request 3 days beam time on IN5 and 3 days beam time on IN4 to probe the crystal field excitations in both $Tb_2Ir_2O_7$ and $Er_2Ir_2O_7$ and the magnetic excitations in the "all-in all-out" magnetic ordered phase in $Tb_2Ir_2O_7$.

EXPERIMENTAL REPORT

EXPERIMENT N° 4-01-1380 INSTRUMENT: IN4C, IN5 & IN6

DATES OF EXPERIMENT IN6 : 29/08/2014 → 01/09/2014, IN4C : 14/11/2014 → 17/11/2014

TITLE: **Magnetic and Crystal Field Excitation Spectra in the Iridate Pyrochlores $Tb_2Ir_2O_7$ and $Er_2Ir_2O_7$**

EXPERIMENTAL TEAM: (names and affiliation)

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Date of report: 14 April 2014

Attention of the condensed matter community was recently attracted by the iridate pyrochlores $R^{3+}{}_2Ir^{4+}{}_2O_7$, where, owing to the competition between spin-orbit, crystal field and electron-electron interactions, the Ir-5d electrons might stabilize unprecedented electronic phases [1, 2]. Furthermore, inherently to the geometric frustration combined with the 4f-5d exchange interactions, these $R^{3+}{}_2Ir^{4+}{}_2O_7$ compounds might display novel magnetic phases. Almost all the members of the series exhibit a metal–insulator transition accompanied by a magnetic transition of the Ir sublattice, with an increase of the transition temperature when one scans the R species from the lightest to heaviest. The high temperature phase is a metal for the lighter R [3]. It was proposed theoretically and demonstrated experimentally, although with strong hypothesis, from μSR and resonant magnetic X-ray diffraction that the Ir sublattice magnetic order might be the so-called "all-in/all out" one [4]. The magnetic moment of the Ir magnetic moment, less than 0.1 μ_B and neutron absorber, is very difficult to probe by neutron diffraction, but possible ordering configurations can be indirectly determined from the magnetic order of the rare-earth sublattice using symmetry arguments [5].
To test this idea, we considered $Y_2Ir_2O_7$, $Tb_2Ir_2O_7$ and $Er_2Ir_2O_7$, which we have investigated by magnetization measurements at the Institut Néel and by neutron diffraction at ISIS [5]. We assumed that the local magnetocrystalline anisotropies associated with the Tb^{3+} and Er^{3+} ions will be of the easy-axis versus easy plane type, in agreement with the opposite signs of the B^2_0 Steven's factors. Thermomagnetic measurements performed according to the ZFC-FC protocol on the three samples showed that a magnetic ordering of the Ir sublattice occurs at 155 K, 140 and 130 K respectively for Y, Er and Tb compounds. In $Er_2Ir_2O_7$, no magnetic signal is detected by powder neutron diffraction down to 2 K, suggesting an absence of magnetic order of the Er sublattice. In $Tb_2Ir_2O_7$, powder neutron diffraction evidenced magnetic peaks below 40 K, the analysis of which indicated that the Tb moments order in the all-in all-out configuration. The inspection of the magnetic intensities suggests that the Tb moments are first polarized in the Ir molecular field before being fully ordered in the same magnetic order by Tb-Tb exchange at 5 K. We rationalized these results by the compatibility or not of the Ir molecular field with the rare-earth magnetocrystalline anisotropy.
The aim of the proposed inelastic neutron scattering experiments was to probe in detail the crystal field (CF) transitions in the Er and Tb compounds as a function of the temperature and to compare their energy spectrum to the Y compound one where these CF transitions should be absent. This step is a prerequisite for a precise understanding the rare-earth-Ir coupling and the magnetic ground states expected for the rare-earth sublattice in these interesting materials. These time-of-flight experiments were performed on polycrystalline samples of $Y_2Ir_2O_7$, $Tb_2Ir_2O_7$ and $Er_2Ir_2O_7$ on the IN4C, IN6 and IN5 (1 day test experiment) spectrometers. The scattering data were collected between 300 and 1.6 K.
The Y compound revealed phonon modes and no signature from the Ir magnetism as expected due to the weakness of the Ir magnetic moment.

In the Tb compound, large non-dispersive excitations were observed at 10 and 35 meV attributed to crystal field transitions. In the low energy region, a Q-modulated quasielastic signal was further split below 20 K into two excitations at 0.8 and 1.5 meV (figures 1, 2 3). This strongly resembles what was observed in the other Tb pyrochlores with Ti and Sn instead of Ir where it was attributed to the splitting of the ground and first excited doublets [7]. In the Er compound (figures 2, 4), a large amount of magnetic Er_2O_3 impurity was present. On the IN4 instrument, the impurity alone, as well as the contaminated $Er_2Ir_2O_7$ sample, were measured to estimate the contribution of the impurity. This was not done on IN6/IN5 by lack of time. Crystal field transitions were observed around 5, 11 and 18 meV on IN4, again in good agreement with the results obtained on the other Er titanate and stanate pyrochlores. On IN6, further crystal field level transitions at higher temperatures were recorded (2, 3, 8 meV) and one mode was observed at 0.45 meV at low temperature, which emerges from the quasielastic signal. The origin of this energy level transition is puzzling since for this Kramer ion, one does not expect the splitting of the ground state doublet by a molecular field.

Crystal field calculations are under way in order to derive from these experiments the crystal field level scheme of the Er and Tb iridate compounds and to relate these findings to their magnetic properties: influence of the Ir molecular field, possible distortion, nature of the low temperature Er ground state.

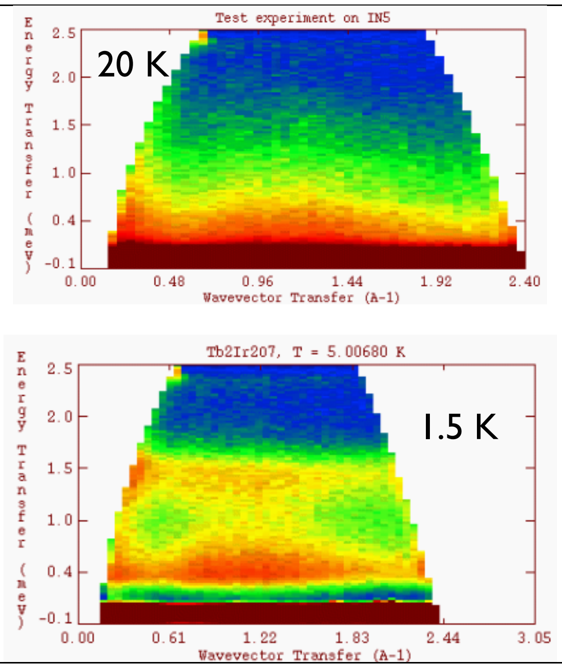

Figure 1: Inelastic scattering intensity as function of wavevector transfer and energy transfer recorded on IN5 for $Tb_2Ir_2O_7$ at 1.5 K (top) and 20 K (bottom)

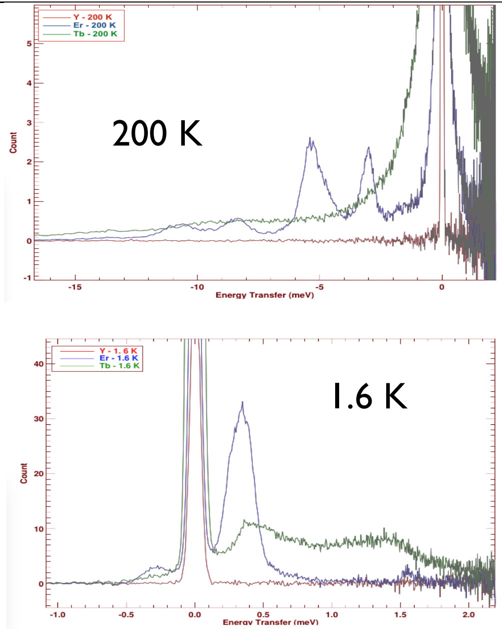

Figure 2: Inelastic scattering intensity as function of energy transfer recorded on IN6 for $Y_2Ir_2O_7$ (in red), $Er_2Ir_2O_7$ (in blue) and $Tb_2Ir_2O_7$ (in green) at 200 K (top) and 1.6 K (bottom)

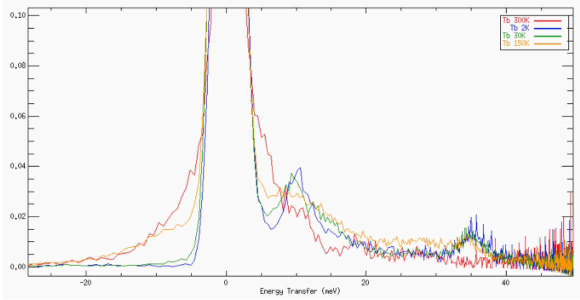

Figure 3: Inelastic scattering intensity as function of energy transfer recorded on IN4 for $Tb_2Ir_2O_7$ at 300 K (in red), 150 K (in yellow), 30 K (in green) and 2 K (in blue)

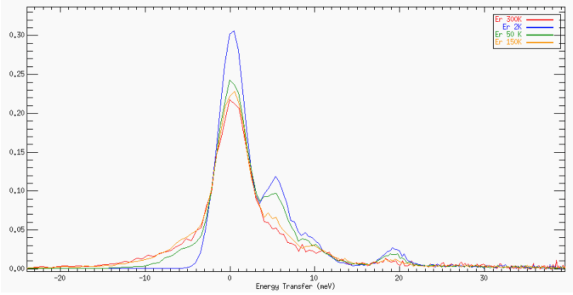

Figure 4: Inelastic scattering intensity as function of energy transfer recorded on IN4 for $Er_2Ir_2O_7$ at 300 K (in red), 150 K (in yellow), 30 K (in green) and 2 K (in blue)

References:

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