Experimental report

31/03/2017

Proposal: 4-01-1478 **Council:** 4/2015

Title: Magnetic and crystal field excitations in the pyrochlore iridates R2Ir2O7 (R=Yb, Dy, Ho, Gd)

Research area: Physics

This proposal is a new proposal

Main proposer: Emilie LEFRANCOIS

Experimental team: Emilie LEFRANCOIS
Virginie SIMONET
Elsa LHOTEL
Rafik BALLOU

Local contacts: Bjorn FAK
Jacques OLLIVIER

Samples: Yb2Ir2O7
Ho2Ir2O7
Dy2Ir2O7
Gd2Ir2O7

Instrument	Requested days	Allocated days	From	To
IN6	0	6	22/10/2015	28/10/2015
IN5	6	0		
IN4	4	6	27/11/2015	03/12/2015

Abstract:

The proposed experiment is part of our systematic experimental studies of the pyrhochlore iridates R2Ir2O7 (R = Rare earth). In these, except for Pr, the Ir-5d electrons exhibit a metal-insulator transition accompanied with a magnetic freezing. This, through f-d exchange, induces an "all-in/all-out" magnetic order of the Tb moments in Tb2Ir2O7. In contrast, the Er moments in Er2Ir2O7 would exhibit spin liquid correlations, suggesting strong relevance of the R3+ magneto-crystalline anisotropy and prompting us to extend our investigations to other species R. We wish here to probe the excitations spectra in Gd2Ir2O7 (where the Gd3+ ions are isotropic), Dy2Ir2O7 and Ho2Ir2O7 (where the Dy3+ and Ho3+ ions should show local easy-axis anisotropy) and Yb2Ir2O7 (where the Yb3+ ions should show local-easy plane anisotropy). A proposal has been submitted on D1B and D4 to determine beforehand the low temperature magnetic phases. It is emphasized that the topic is hot, as a unique example of correlated topological insulators coupled to a possibly frustrated R magnetism, and that it will be harmful to postpone the submission of the inelastic proposal after the elastic experiment.

EXPERIMENTAL REPORT

EXPERIMENT N° 4-01-1478 INSTRUMENT: IN4-IN6

DATES OF EXPERIMENT : IN4: 27/11/2015 → 03/12/2017 IN6: 22/10/2015 → 28/10/2015

TITLE: Magnetic and crystal field excitations in the pyrochlore iridates R2Ir2O7 (R=Yb, Dy, Ho, Gd)

EXPERIMENTAL TEAM: (names and affiliation)

Emilie LEFRANCOIS and Laurent CHAPON, ILL, Grenoble,

Rafik BALLOU, Pascal LEJAY, Virginie SIMONET, Elsa LHOTEL, Institut Néel, CNRS, Grenoble

LOCAL CONTACT: Bjorn FAK, Jacques OLLIVIER

Date of report: 30 mars 2017

Pyrochlore iridates of formula $R_2Ir_2O_7$, feature a new family of magnetic materials with fascinating properties as concerns the role of the spin-orbit coupling in the stabilization of novel electronic phases [1], but also from the point of view of magnetic frustration. Indeed, in these materials, both the rare-earth and the iridium ions occupy a pyrochlore lattice, made of corner-sharing tetrahedra and prone to frustration. Moreover, as demonstrated by our previous study on the Tb and Er compounds [2], as well as by other research teams, the Ir^{4+} magnetic moments order at rather high temperature (above 120 K for all rare-earths smaller than Nd) in the all-in all-out configuration, that is to say with all spins pointing inward or outward of each tetrahedron. This magnetic arrangement produces on the rare-earth sublattice site a local magnetic field oriented locally along the <111> directions that also follows an all-in all-out configuration. This staggered field can go along with the single-ion anisotropy of the rare-earth and with the magnetic correlations promoted by the interactions between the rare-earths. This is the case of the Tb compound, which displays an easy-axis of anisotropy along the <111> local directions and presents Tb-Tb antiferromagnetic interactions [2]. The local field can however rather compete with the single-ion anisotropy (case of the Er with an easy-plane of anisotropy [2]) and/or with the R-R interactions. This is the case of the Ho where, due to the R-R ferromagnetic interactions, the presence of the local field leads to a spectacular fragmentation of the Ising-like magnetic moment into a persistently fluctuating part (Coulomb phase) and an ordered part (magnetic charge crystal) [3,4].

The aim of the proposed inelastic neutron scattering experiments was to probe in detail the crystal field (CF) transitions in the Yb, Dy and Ho compounds as a function of the temperature. This step is a prerequisite for a precise understanding of the magnetic ground states expected to be exotic for the rare-earth sublattice in these interesting materials. In addition, the low energy spectrum measured in the same compounds plus the Gd one (where no CF transitions are expected) allows to access the magnetic excitations associated with the low temperature magnetic phase and thus to the rare-earth-Ir coupling. These time-of-flight experiments were performed on polycrystalline samples of Yb2Ir2O7, Dy2Ir2O7, Ho2Ir2O7 and Gd2Ir2O7 (with isotopic ^{156}Gd) on the IN4 and IN6 spectrometers. The scattering data were collected between 300 and 1.5 K.

On IN4, we could measure the crystal field excitations (see figure 1) which are consistent with the ones observed in the other pyrochlore compounds, where the Ir ion is replaced by Ti or Sn. Our crystal field analysis gives a similar ground state doublet as titanates and stannates, with a strong Ising anisotropy for Dy and Ho, and an easy plane anisotropy for the Yb (See Ref [3] for the Ho analysis).

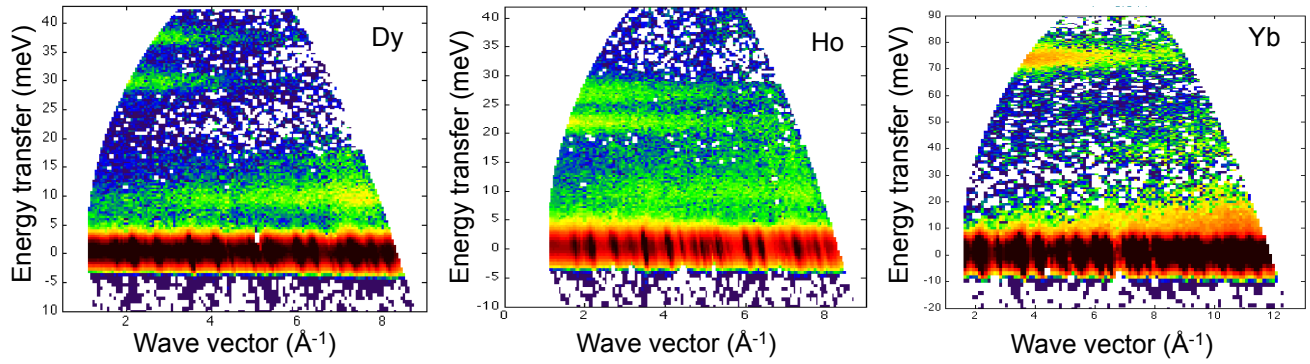

Figure 1: Inelastic scattering intensity as function of wavevector transfer and energy transfer recorded on IN4 at 1.5 K for Dy2Ir2O7 (λ=1.3 Å), Ho2Ir2O7 (λ=1.3 Å) and Yb2Ir2O7 (λ=0.9 Å).

In the low energy region measured on IN6 (see Figure 2), a splitting of the ground state Ising doublet is observed on the Ho compound, which is due to the Ir molecular field. This splitting is not observed on the Dy one due to its Kramers nature.

In Yb2Ir2O7, a large excitation is observed at low temperature, which is certainly associated with the ferromagnetic Yb-Yb correlations, since it resembles the energy spectrum observed in Yb2Sn2O7 [5].

In Gd2Ir2O7, below 50 K, we observed an excitation, with a Q-modulated signal and an energy gap of about 0.3 meV. Our analysis shows that the gap can be explained by the interplay between the Ir molecular field, and the small easy plane anisotropy of the Gd ion, which is due to admixture with the higher multiplet states. The understanding of the modulation, which features a collective excitation, is not achieved at the moment.

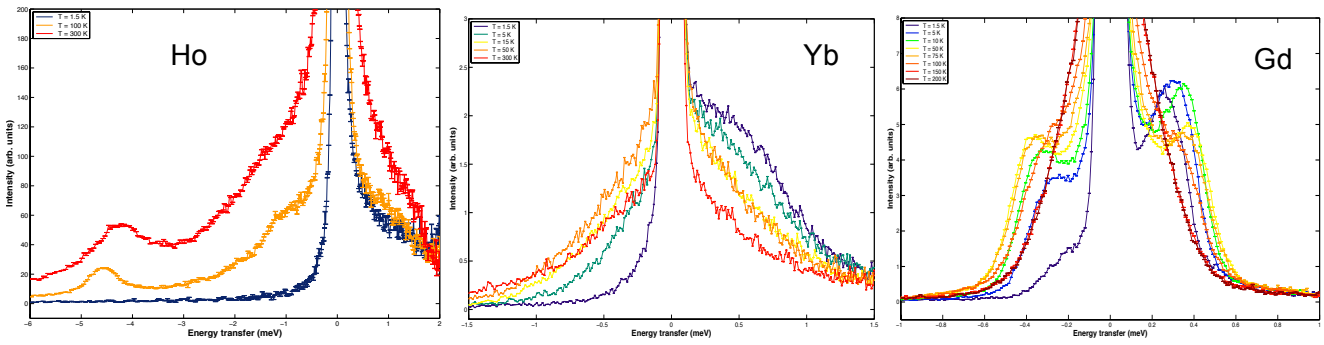

Figure 1: Inelastic scattering intensity as function of energy transfer recorded on IN6 at λ=5.1 Å at several temperatures between 1.5 K and 300 K for Ho2Ir2O7 and Yb2Ir2O7, and between1.5 K and 200K for Gd2Ir2O7

References:

[1] Wan *et al.*, Phys. Rev. B 83, 205101 (2011).

[2] Lefrançois *et al.* Phys. Rev. Lett. 114, 247202 (2015).

[3] Lefrançois *et al.* arXiv:1702.02864.

[4] Brooks-Bartlett *et al.*, Phys. Rev. X 4, 011007 (2014).

[5] Dun *et al.*, Phys. Rev. B 87, 134408 (2013).