Experimental report

15/09/2016

Proposal: **5-31-2406** **Council:** 4/2015

Title: Investigation of the magnetic structure of the pyrochlore iridates R2Ir2O7 (R=Yb, Ho, Dy and Gd)

Research area: Physics

This proposal is a new proposal

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Samples: Yb2Ir2O7
Ho2Ir2O7
Dy2Ir2O7
Gd2Ir2O7

Instrument	Requested days	Allocated days	From	To
D1B	4	4	29/10/2015	02/11/2015
D4	4	4	15/10/2015	19/10/2015

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 low temperature magnetic phase of 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 for inelastic scattering has been submitted on IN5 and IN4 to also probe the excitation spectra of possible magnetic ordering and the crystal field spectra.

EXPERIMENTAL REPORT

EXPERIMENT N° 5-31-2406 INSTRUMENT: D4 and D1B

DATES OF EXPERIMENT : D4: 15/10/2015 → 19/10/2015 and D1B: 29/10/2015 → 02/11/2015

TITLE: **Investigation of the magnetic structure of the pyrochlore iridates $R_2Ir_2O_7$ (R = Yb, Ho, Dy and Gd)**

EXPERIMENTAL TEAM: (names and affiliation)

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Date of report: 15.09.2016

Attention of the community of condensed matter 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] and, inherently to the geometric frustration combined with the 4f-5d exchange interactions, the R-4f electrons might display novel magnetic phases. Almost all the members of the series exhibit a metal–insulator transition accompanied by a magnetic ordering, 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. It is a semimetal then a semiconductor for increasingly heavy R [3]. The Pr-based compound remains metallic at all temperatures and shows a spontaneous anomalous Hall effect, suggesting that it might stabilize a chiral metallic spin liquid. The Nd-based compound displays a metal–insulator transition. It was argued from powder neutron diffraction experiments that its magnetic order might be the so-called "all-in/all-out" (AIAO) (see figure 2) in consistency with the topological nature of the insulating phase [4]. The magnetic arrangement of the Ir moments was actually inferred indirectly and this was questioned from muon spin relaxation/ rotation experiments [5]. The magnetic moment of Ir itself is very difficult to detect, but it can be probed through the coupling between the Iridium and the rare-earth sublattice using rare-earth with different types of anisotropy [6]. We have shown that for the Terbium based compound (local easy axis of anisotropy) the Tb sublattice orders in the AIAO configuration, the order being induced through the molecular field produced by the Ir sublattice which is parallel to the easy-axis. On the contrary for the Erbium based compound (local easy plane of anisotropy), we have found a disordered phase, most likely a spin-liquid like state, the anisotropy competing with the molecular field.

We studied $Ho_2Ir_2O_7$, $Yb_2Ir_2O_7$, $Dy_2Ir_2O_7$ and $Gd_2Ir_2O_7$. These compounds have been investigated only partially so far [3,6]. It is expected that the local magnetocrystalline anisotropies associated with the rare-earth ions (easy axis for Ho^{3+} and Dy^{3+}, easy plane for Yb^{3+} and isotropic for Gd^{3+}) being different will lead to drastically different behaviors as we have observed between $Tb_2Ir_2O_7$ and $Er_2Ir_2O_7$ [7]. A magnetic freezing is evidenced in the ZFC-FC magnetization measurements at 145 K in $Yb_2Ir_2O_7$, 135 K in $Ho_2Ir_2O_7$ and at 130 K in $Dy_2Ir_2O_7$. An all-in/all-out magnetic order cannot give rise to uniform magnetization and the thermomagnetic signals arise from defects [6], actually revealing only Iridium magnetic freezing. An elastic neutron scattering experiment is necessary to microscopically determine whether the R^{3+} moments order at low temperature or remain in a disordered state.

The aim of the proposed experiments is to determine the low temperature magnetic phases of the different samples. Assuming that the Iridium orders magnetically in the all-in/all-out configuration within the whole series, we expect in the case of $Ho_2Ir_2O_7$, $Dy_2Ir_2O_7$ and $Gd_2Ir_2O_7$ that the rare-earth sublattice will order also in the all-in/all-out configuration through the rare-earth/Ir magnetic exchange. Indeed the Ir moments then will produce a molecular field parallel to the local rare-earth 3-fold axis, which is compatible with the local easy axis magnetocrystalline anisotropy of Ho^{3+} and Dy^{3+} ions and the isotropy of the Gd^{3+} ions. On the other hand, for $Yb_2Ir_2O_7$, we expect a spin-liquid behavior as it presents an easy plane of anisotropy perpendicular to the Ir molecular field.

The measurements show indeed the presence of magnetic Bragg peaks corresponding to an all-in/all-out magnetic arrangement of the rare-earth magnetic moments for $Ho_2Ir_2O_7$ and $Dy_2Ir_2O_7$ (see Fig. 1) while for $Yb_2Ir_2O_7$ no additional signal was observed suggesting the absence of long range order down to 1.5 K (see Fig. 2). These behaviors are consistent with the previous results on the Er and Tb based compounds and the expected anisotropy of these rare earth elements.

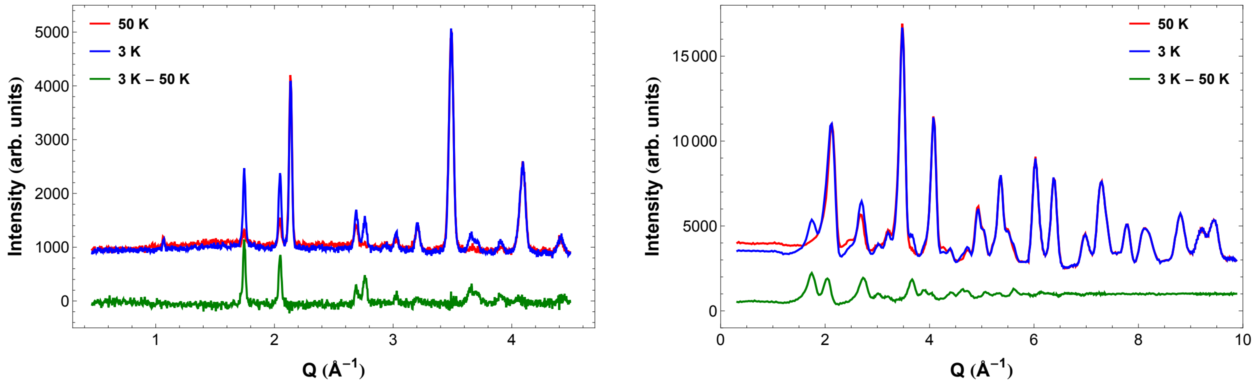

Figure 1: (left) Neutron diffractograms recorded for $Ho_2Ir_2O_7$ on D1B (left) and for $Dy_2Ir_2O_7$ on D4c (right) at 3 K (blue) and 50 K (red). The difference between the 1.5 K and 50 K data is shown in green.

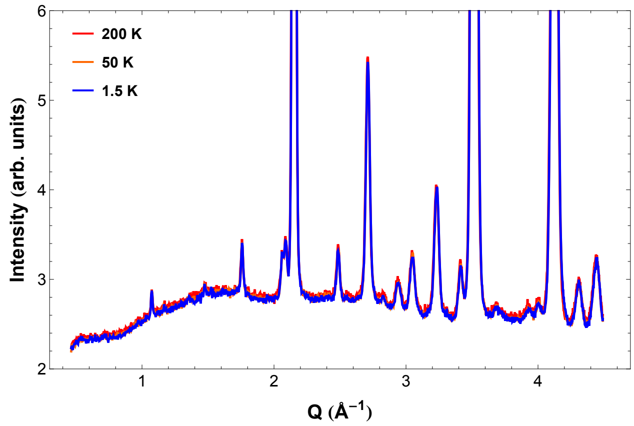

Figure 2: Neutron diffratograms recorded for $Yb_2Ir_2O_7$ at 1.5 (blue), 50 (orange) and 200 K (red).

For $Gd_2Ir_2O_7$, the diffraction measurements were first performed on a compound with natural Gd on D4c. Magnetic Bragg peaks associated to an all-in/all-out ordering of Gd moments are observed as expected for isotopic spins. However an additional signal is observed around 1.2 Å-1 (see left part of Fig. 3). This signal is not resolution limited and rather seems to arise from short range correlations. This result was then confirm by diffraction measurements performed on D1B using a compound with isotopic Gd (see right part of Fig. 3). Thus in $Gd_2Ir_2O_7$, the coexistence of both short range and long range correlations is observed. Theoretical calculations and additional measurements are under way in order to understand this magnetic behavior.

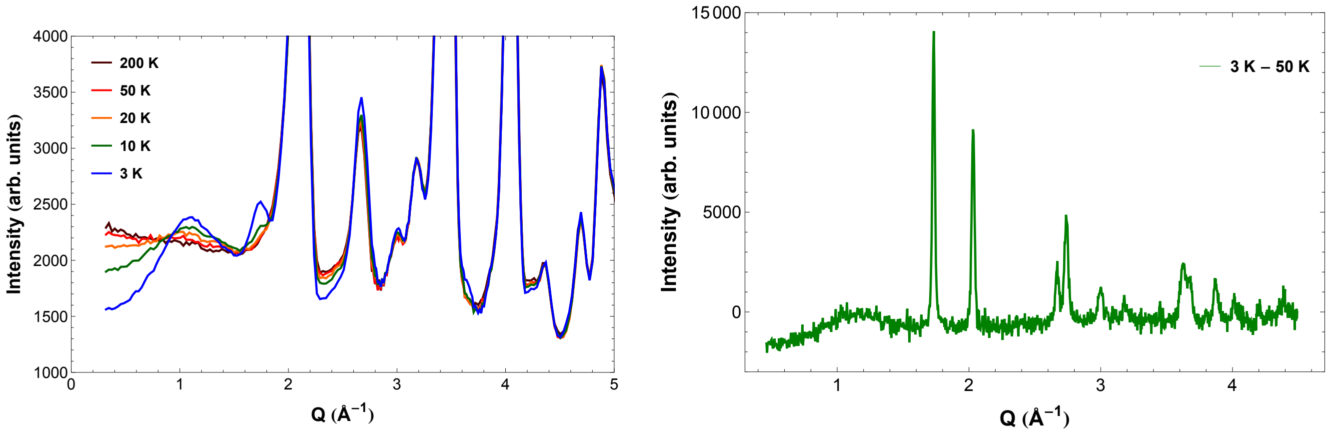

Figure 3: (left) Neutron diffratograms recorded for $Gd_2Ir_2O_7$ between 3 K and 50 K on D4c. (right) Difference between the diffractograms recorded at 3 and 50 K on D1B.

References:

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