Experimental report

10/10/2024

Proposal: **5-31-2926** **Council:** 10/2022

Title: Magnetic phase diagram under pressure of Ho pyrochlore iridate

Research area: Physics

This proposal is a continuation of 5-31-2800

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Samples: $Ho_2Ir_2O_7$

Instrument	Requested days	Allocated days	From	To
D20	3	3	08/04/2023	11/04/2023

Abstract:

In pyrochlore iridates (formula $R_2Ir_2O_7$), the iridium magnetic moments apply a molecular field on the rare-earth ions, resulting in a wide variety of magnetic phases. Recently, a theoretical phase diagram was computed, showing that by varying the Ir molecular field and the rare-earth interactions, one can go from an antiferromagnetic state to a fragmented phase (where the magnetic moment fragments into an ordered antiferromagnetic phase and a fluctuating phase) and to a spin ice state. We have started to explore this phase diagram experimentally by measuring the magnetic structure under pressure in $Ho_2Ir_2O_7$ up to 10 GPa. This experiment showed that pressure is a relevant tool to probe the phase diagram and weakens the fragmented ground state stabilized at ambient pressure. However, the applied pressure was not high enough to reach the phase boundary. The objective of the present experiment is thus to increase the pressure further up to 20GPa to reach this phase boundary and compare with the theoeretical predictions.

Magnetic phase diagram under pressure of Ho pyrochlore iridates. Diffraction in PE cell at D20.

Aim of the experiment: the aim of this experiment was to probe the influence of isostatic pressure on the magnetic ground state of $Ho_2Ir_2O_7$, hence to explore the (P, T) phase diagram experimentally. Indeed, we expect that applying pressure will change magnetic exchange paths, and in turn the magnetic structure. At ambient pressure, the Ho magnetic moment in $Ho_2Ir_2O_7$ orders in an “all in – all out” structure when decreasing the temperature. The ordered moment saturates at half of the total moment, which is the signature of the stabilization of a monopole crystal (“fragmented”) structure. A rich phase diagram has been proposed theroretically around this unconventional state. The pursued idea is that pressure could modifiy the different coupling parameters and induce a transition towards a different ground state. This experiment is the continuation of a previous one, carried out on the same instrument, where we could reach 10 GPa and measure the All-in All-out magnetic moment as a function of T and P.

Experimental conditions: The measurements were carried out on a powder sample at D20, in April 2023. We used a PE pressure cell, allowing one to apply pressure up to 20 GPa. The sample was loaded in a gasket (to minimize the neutron background) and soaked with a few drops of a (deuterated) ethanol-methanol mixture. Two hemispheres were prepared and then glued together by applying a small pressure of about 80 bars. A tiny “pig tail” lead sample was added in these hemispheres to directly measure *in situ* the pressure. D20 was operated with a PG002 monochromator and $\lambda = 2.4$ Å. We recorded diffractograms on cooling, as well as at fixed temperature of 5, 7, 10, 15 and 80K. To save time, the cryostat was cooled down very quickly from 300 to 80K using liquid nitrogen, which was then removed and pushed away with dry air.

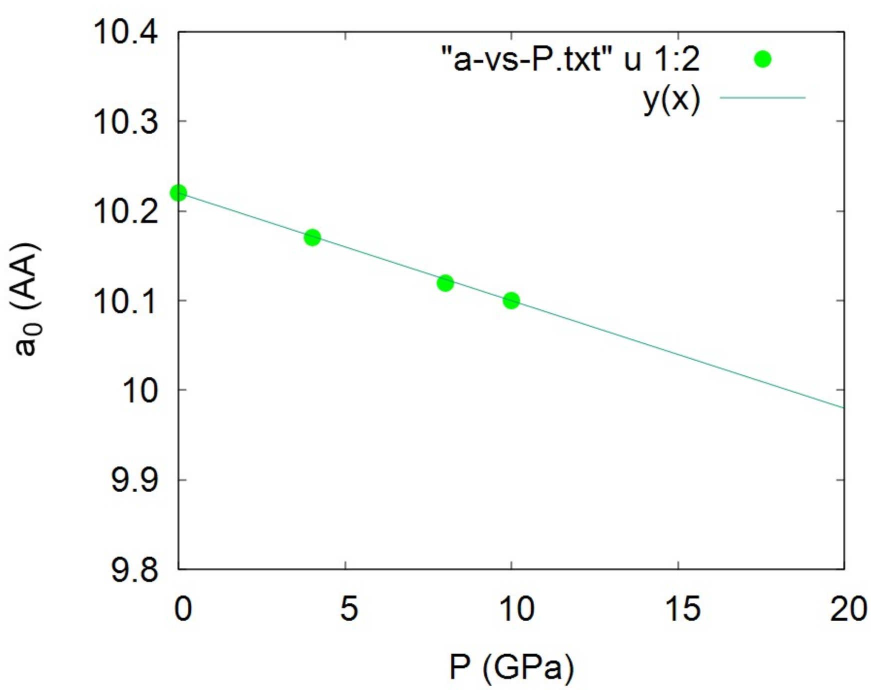

FIG. 1. Evolution of the $Ho_2Ir_2O_7$ lattice parameter as a function of pressure.

Our first attempt during this second run was unsuccessfull as we broke the diamonds. The second and third tries were OK, reaching 7 and about 11 GPa. During the fourth one, increasing the pressure up to 20 GPa, the sample collapsed again and we decided to stop.

The evolution of the lattice parameter vs pressure is shown in Figure 1. The data prove however quite difficult to refine with Fullprof, but the analysis is still under progress. Raw data are shown in Figure 2. At ambient pressure and low temperature, the observed magnetic structure is "all in – all out", characterized by a k=0 propagation vector [1]. Relevant magnetic Bragg peaks of the structure are especially (220) and (331). Our preliminary analysis consists in fitting the (220) profile at the different temperatures and adjust the integrated intensity according to $A + Bm^2$, where A, B are coefficients that can be determined by analytical calculations. Both are scaled to the nuclear response using Fullprof along with the 80 K data. This fitting suggest that pressure tends to reduce the Ho ordered moment from 5 down to 3.4 and 3.2 a 11 GPa. These values are consistent with the first experiment. Unfortunately, the evolution at larger pressure remains unknown.

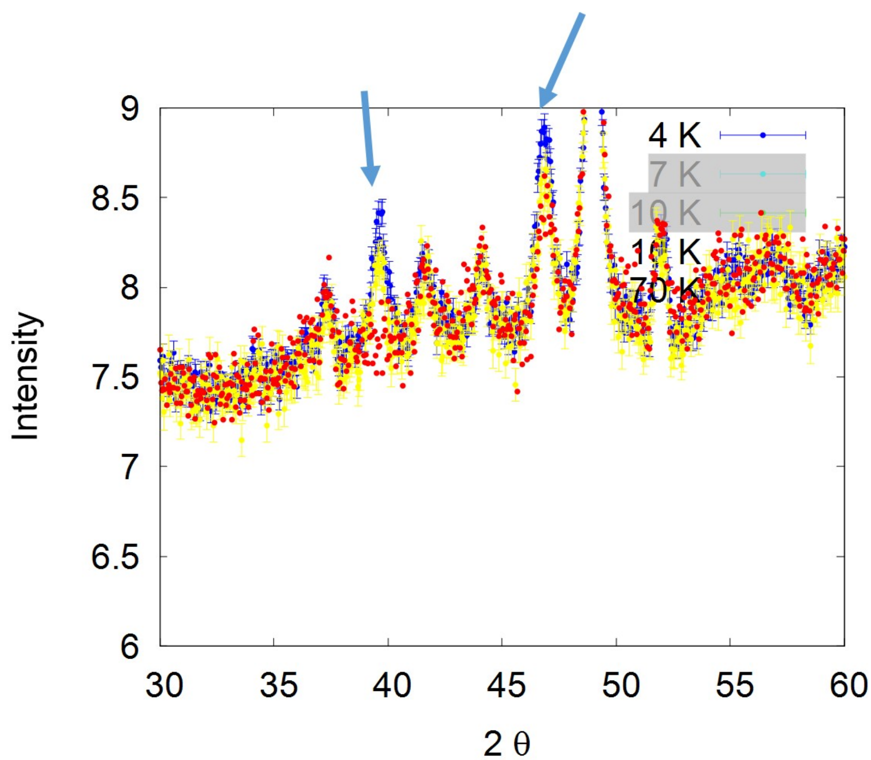

FIG. 2. Raw diffractograms taken at 7 GPa.

References: [1] Lefrançois et al., Nature Commun. 8, 209 (2017).