Experimental report

27/05/2022

Proposal: **5-31-2858** **Council:** 4/2021

Title: Relationship of magnetic ordering and crystal structure in lanthanideferromagnets Gd, Dy, and Ho at high pressures

Research area: Physics

This proposal is a new proposal

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Samples: 160Gd
160 Dy
Nat Ho

Instrument	Requested days	Allocated days	From	To
D20	6	0		
D1B	6	2	28/06/2021	30/06/2021

Abstract:

The ferromagnetism of lanthanoid metals is modified when the crystal structure changes. All 4f lanthanide ferromagnets, from Gd to Tm, have an hcp structure at ambient pressure, and exhibit the structural transformations in the sequence hcp -> Sm-type -> double-hcp (dhcp) -> fcc -> trigonal under pressure.
The magnetic structures determined by neutron diffraction experiments performed in Tb are in agreement with our high sensitivity a.c. magnetization experiments. However, it is not the case for the Dy, Ho and Gd compounds.
In order to shed light in this discrepancy, we propose to perform neutron powder diffraction experiments in Dy, Ho and Gd - by using the 160Gd and 160Dy isotopes for which the neutron absorption will not be a major problem- at high pressures ($P < 100$kbar).
To accomplish the experiment, we apply for 3 beamtime days at the D20 instrument in its maximum flux configuration

Report of experiment 5-31-2858:

Title: Relationship of magnetic ordering and crystal structure in lanthanide ferromagnets Ho, Tb at high pressures

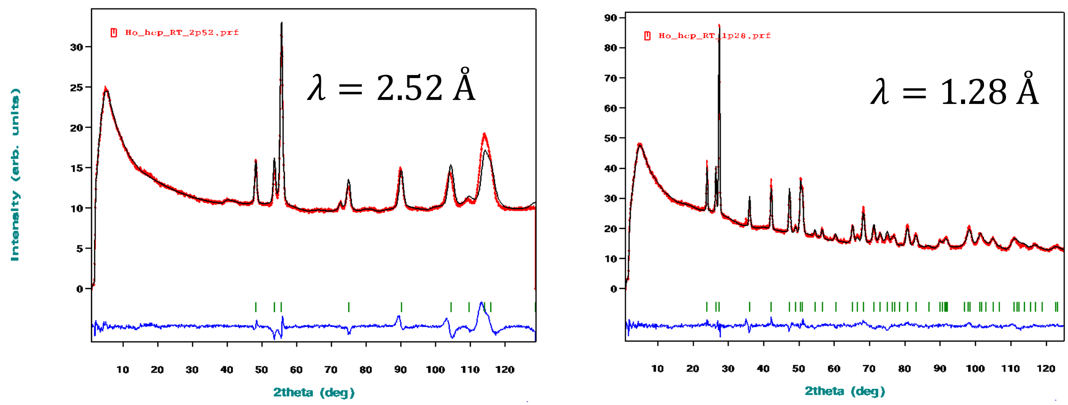
Motivation:

Ferromagnetic metals have been important systems in condensed matter physics from the viewpoint of magnetism originating from itinerant electrons. In the 4f lanthanide series, there are six ferromagnetic (FM) elements Gd, Tb, Dy, Ho, Er, and Tm. This ferromagnetism is modified when the crystal structure changes. All 4f lanthanide ferromagnets, from Gd to Tm, have an hcp structure at ambient pressure, and exhibit the structural transformations in the sequence hcp → Sm-type → double-hcp (dhcp) → fcc → trigonal under pressure.
The variation of the magnetic properties with the structural transformations in 4f ferromagnetic metals has been reported by magnetic measurements [1-4], electrical resistivity [5-10], neutron diffraction [6, 11, 12], and Mossbauer spectroscopy [13]. Recently, neutron diffraction experiments for Tb [6], Dy [11], and Ho [12] have been reported at high pressures. In Dy and Ho, peaks coming from the magnetic order were observed at pressures above Pc in disagreement with previous magnetic measurements [2, 3].
The neutron diffraction experiments performed in Tb are in agreement with our high sensitivity AC magnetization experiments. However, it is not the case for the Dy and Ho compounds. For both, the magnetic peaks observed at high-pressure phases in the neutron diffraction experiments are not consistent with the magnetic anomalies tracing the FM and HM orders observed in our magnetic measurements. The presence or not, of new magnetic incommensurate peaks (or signals) in the low q region of the diffractograms and its evolution with the temperature will allow to solve the discrepancies.

Experiment 5-31-2858:

This experiment was performed at D1B in ILL (Grenoble, France) from 28/06/21 to 30/06/21 (48 hours).
Although the first idea was to perform measurements on both 160Gd isotope and Ho, the former one had a small impurity of 157Gd (less than 0.4%), which made the absorption so high that no feasible experiment could be carried out on this sample.
Several measurements were performed on Ho powder, which can be summarized as follows:

- Measurements at atmospheric pressure (AP) and room temperature (RT) with λ = 1.28 and 2.52 Å were taken with the Ho sample inside a vanadium holder.
- Measurements at 8 GPa with λ = 2.52 Å. For these measurements the powder was introduced inside the Paris-Edinburgh (PE) pressure cell, then prepared in the gasket using a 4:1 ethanol-methanol mix as pressure transmitter medium. Once the sample was inside, a pressure of 550 bar was applied to the PE cell, which corresponded to 8 GPa inside. Then the system was cooled using liquid nitrogen and helium from RT to 5 K. At this point a longer measurement was performed for a fixed temperature (5 K) and then data were taken as the system was heated back to RT

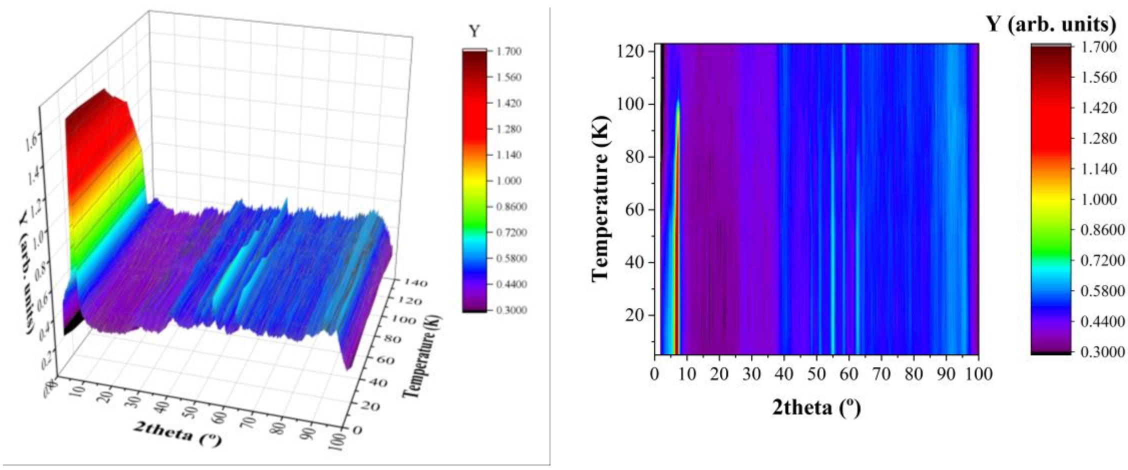


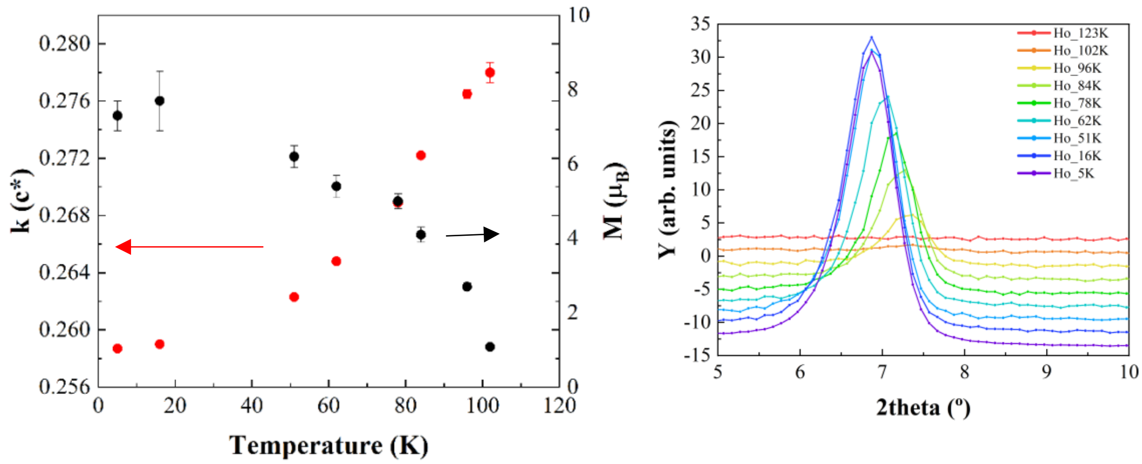
As it is shown above, the peaks observed in the diffraction patterns at AP and RT can be indexed by the space group $P6_3/mmc$ (No. 194). This crystal structure corresponds with a hcp structure where the Ho atom is in the Wyckoff position 2c (1/3, 2/3, 1/4). The cell parameters are $a = b = 3.5612(2)$; $c = 5.5895(4)$; $\alpha = \beta = 90º$; $\gamma = 120º$.

In the case of the measurements at 8 GPa and 5 K, the intensity of the nuclear peaks is greatly suppressed, as the quantity of sample under pressure which is irradiated by the neutron beam is much smaller.
Nevertheless, the peaks can still be indexed by the same hcp structure (space group $P6_3/mmc$ (No. 194)), where the cell parameters are fitted to: $a = b = 3.395(1)$; $c = 5.330(3)$; $\alpha = \beta = 90º$; $\gamma = 120º$.

Regarding the magnetic structure, it is possible to observe a high intensity magnetic peak at $2\theta = 7º$ which can be indexed with a propagation vector $\vec{k} = (0, 0, 0.2586(1))$ (in units of c^*), in agreement with [12].

From the temperature ramps obtained at 8 GPa as the system was being heated from 5 K to RT, we can observe that the onset of this helical magnetic ordering occurs around $T_N = 99(3)$ K. We can plot also the evolution of the propagation vector module as well as the modulus of the Ho magnetic moment. As the temperature decreases, the magnetic moment increases from 0 to 7.3(4) μ_B, while the propagation vector decreases its value from 0.2780(7) to 0.2587(1), implying a longer period of the helix at lower temperatures.





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