Experimental report

21/05/2024

Proposal:	**5-31-2947**	**Council:**	10/2022

Title: Relationship of magnetic ordering and crystal structure in the ferromagnet Tm at high pressures

Research area: Physics

This proposal is a new proposal

Main proposer: **Javier CAMPO RUIZ**

Experimental team:
Francisco RUBIO
Javier CAMPO RUIZ
Yusuke KOSAKA
Masaki MITO

Local contacts:
Jose Alberto RODRIGUEZ VELAMAZAN
Ines PUENTE ORENCH

Samples: Tm -Thulium

Instrument	Requested days	Allocated days	From	To
D20	3	3	23/06/2023	26/06/2023
D1B	3	0		

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.

Recently we have also performed high sensitivity a.c. magnetization experiments for Er and Tm. It is noted that Er has both incommensurate (HM) and FM orders at least 30 GPa, whereas Tm has the disappearance of a.c. magnetization anomaly due to the HM ordering near the hcp-Sm structural transition. The experimental results suggest that the HM order is more stable than the FM one for Tb, and Ho, whereas in Tm, the FM is more stable than the HM order. This may be an general difference between light 4f lanthanide FM elements (Tb-Ho) and heavy one (T m)

There has been no neutron diffraction data for Tm at high pressures so far. In order to seed light in this discrepancy between Tb-Ho and Tm, we propose to perform neutron powder diffraction experiments in Tm at high pressures (P < 150Kbar)

To accomplish the experiment, we apply for 3 beamtime days at the D20 (or D1B)instrument.

5-31-2947

Relationship of magnetic ordering and crystal structure in the ferromagnet Tm at high pressures

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. Its ferromagnetism is explained by the RKKY interaction among localized moments of the f-orbital electrons mediated by the conduction electrons. The spatially damped oscillation of the conduction electron spin polarization is responsible of the competition between the FM and antiferromagnetic correlations, which often results in an incommensurate magnetic structure (HM). Hereafter the magnetic transition temperatures between the FM and HM states and the transition temperatures between the HM and paramagnetic states are denoted as T_C and T_N, respectively.

The aforementioned 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, as shown in Fig. 1

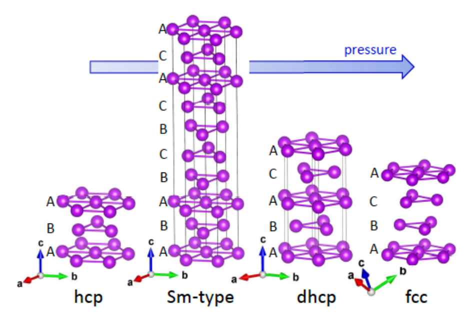

Fig 1. Structural transformation in 4f lanthanide ferromagnets Gd-Tm under pressure from hcp→Sm-type→ double-hcp (dhcp) → fcc structure

The variation of the magnetic properties with the structural transformations in 4f ferromagnetic metals has been reported by magnetic measurements, electrical resistivity, neutron diffraction, and Mossbauer spectroscopy. The previous magnetic measurements on Gd-Ho series under pressure are reviewed by employing high sensitivity magnetic measurements using a diamond anvil cell (DAC) developed by our group. The Figure 2 shows the pressure regions in which the HM and FM magnetizations appear together with the change in the crystal structure. Given these results, the FM order in Gd, Tb, Dy, and Ho can survive stably in the domains with the hcp structure.

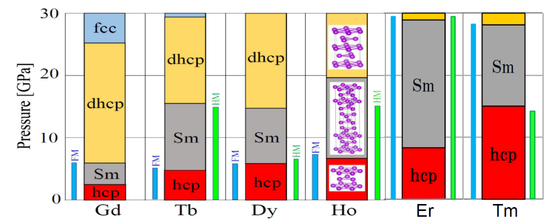

Fig 2. The relationship between the critical pressure Pc for the disappearance of both FM and HM magnetizations and the change in the crystal structure

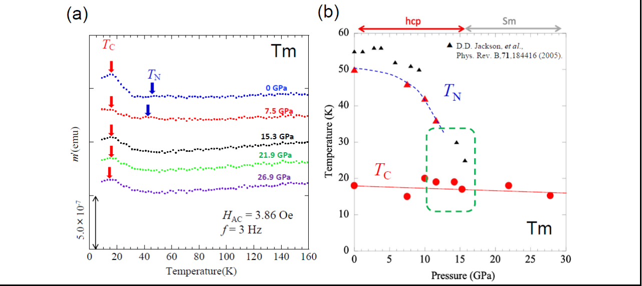

Fig 3. Ac magnetization of Tm as a function of temperature (a) and pressure dependence of transition temperatures for Tm (b). Our work is referred as Mito *et al.*

Recently we have also performed high sensitivity a.c. magnetization experiments for Er and Tm. It is noted that Er has both HM and FM orders at least 30 GPa, whereas Tm has the disappearance of a.c. magnetization anomaly due to the HM ordering near the hcp-Sm structural transition as shown in Fig.3. The experimental results suggest that the HM order is more stable than the FM one for Tb, and Ho, whereas in Tm, the FM is more stable than the HM order. This may be a general difference between light 4f lanthanide FM elements (Tb-Ho) and heavy one (Tm).

There has been no neutron diffraction experiment for Tm at high pressure so far. In order to seed light in this discrepancy between Tb-Ho and Tm, we propose to perform neutron powder diffraction experiments in Tm at high pressures (P < 150Kbar). The presence or not, of new magnetic incommensurate peaks (or signals) in the diffractograms at low **q** and its evolution with the temperature will allow to solve the discrepancies.

Experiment details.

We collected good data at several pressures (5.5, 9 and 15 GPa) and now we are analyzing the data. Here we describe the sequence of the experiments done.

We employed a sample of Tm, with size <150 micrometer inmersed in Fluorinet FC70.
Tm powder sample in double toroidal PE cell. Some pressure (74 bar) to seal the gasket
Acquisitions scanning in 2th from -8 to -5 deg. step 0.05 deg. (61 steps)
240K room P (74 bar). 180 s/step (interrupted after ~1h).
We can see peaks of Pb(1 1 1) at 2th=50 deg and the strongest peak of Tm (1 0 1) at 2th=53.4 deg
Slowly increasing pressure

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----------------------------------------------------------------------------------------------
-------------------------PRESSURE 500 bar = 5.5(1) GPa-----------------------
----------------------------------------------------------------------------------------------
T=80K -------------------------
~ 80K 500 bar = 5.5(1) GPa 2 m/step (2h).
~ 80K 500 bar = 5.5(1) GPa 2 m/step (2h).
cooling down to 30K-------------------------
~ cooling to 30K 500 bar = 5.5(1) GPa 20 sec/step (20min).
T=35 K -------------------------
~ 35K 500 bar = 5.5(1) GPa 2 m/step (2h).
~ 32K 500 bar = 5.5(1) GPa 2 m/step (2h).
~ 32K 500 bar = 5.5(1) GPa 2 m/step (2h).
cooling down to 5K-------------------------
~ cooling to 5K 500 bar = 5.5(1) GPa 20 sec/step (20min).
T=5K -------------------------
~ 5K 500 bar = 5.5(1) GPa 2 m/step (2h).
~ 5K 500 bar = 5.5(1) GPa 2 m/step (2h).
-------------------------Saturday 24 June 2023-------------------------
warm up to 300K ( 8h15m AM)
~ warm up to 300 K 500 bar = 5.5(1) GPa 20 sec/step (20min).
~ warm up to 300 K 500 bar = 5.5(1) GPa 20 sec/step (20min).
T=277K -------------------------
start long adcquisiton at T=277 during the warm up (starting at 15h00)
~ 277 K 500 bar = 5.5(1) GPa 2 m/step (2h). ( T_ini=277, T_fin=300k)
----------------------------------------------------------------------------------------------
-------------------------PRESSURE 650 bar = 9.2(2) GPa-----------------------
----------------------------------------------------------------------------------------------
T=300 K -------------------------
Changing the pressure to 9 GPa 17h15 m.
cooling to 80 (Tsample start 90K) 650 bar  9.2(2) GPa   20s/step (20 min)
T=80K -------------------------
80K 650 bar  9.2(2) GPa   2min/step (20 min)
80K 650 bar  9.2(2) GPa   2min/step (20 min)
T=25 K -------------------------
25K 650 bar  9.2(2) GPa   2min/step (20 min)
25K 650 bar  9.2(2) GPa   2min/step (20 min)
-------------------------Sunday 25 June 2023-------------------------
cooling down to 5K-------------------------
T=5K -------------------------
5K 650 bar  9.2(2) GPa   2min/step (20 min)
5K 650 bar  9.2(2) GPa   2min/step (20 min)
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WARM UP TO 300 K--------------------------
warming up to 250 K 650 bar 9.2(2) GPa   20s/step (20 min) Ist sample temp 242K

-------------------------PRESSURE 900 bar = 15(?) GPa-----------------------
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T=80K --------------------------
80K 900 bar 13.4 GPa   2min/step (2 horas)
80K 900 bar 13.4 GPa   2min/step (2 horas)
-------------------26/06/2023
cooling down to 5K--------------------------
cooling to 5 900 bar 15 GPa   20s/step (20 min)
T=5 K --------------------------
5K 900 bar 15 GPa   2min/step (2 h)
5K 900 bar 15 GPa   2min/step (2h)