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## **Report:**

Understanding the interplay of rare-earth (*R*) local-moment magnetism and 3*d* transition metal itinerant magnetism is of fundamental physical interest and may help in the design of more efficient permanent magnets. The tetragonal  $RFe_4Al_8$  compounds are well suited for studying this interplay because of simple symmetry conditions and because the interaction between the two magnetic sublattices is rather weak. In an x-ray resonant magnetic scattering (XRMS) experiment on DyFe<sub>4</sub>Al<sub>8</sub> [1], dramatic differences were observed in the temperature *T* dependence of the resonances at the Dy  $L_2$  and  $L_3$  absorption edge and attributed to a Dy 5*d* polarization induced by the Fe moments at high *T*, whereas the Dy 4*f* moments order only at low *T*.

We have grown single crystals of GdFe<sub>4</sub>Al<sub>8</sub>, with composition very close to the ideal "148", and characterized them with magnetization, electrical transport, and specific heat measurements, as well as with a prelimary XRMS experiment performed at the Advanced Photon Source, Argonne [2]. In addition to antiferromagnetic order below  $T_N \sim 155$  K, two low *T* transitions at  $T_1 \sim 21$  K and  $T_2 \sim 27$  K were identified. Measuring at the Gd  $L_2$  edge, we found satellite reflections corresponding to an incommensurate propagation vector ( $\tau \tau 0$ ) with  $\tau$  varying between 0.06 and 0.14 as a function of *T*. Similar to the DyFe<sub>4</sub>Al<sub>8</sub> experiment, we interpret this as a polarization of the Gd 5*d* bands induced by Fe moment order. The Gd 4*f* moments order only below  $T_2$  (as deduced from specific heat), with a ferromagnetic component (deduced from magnetization and confirmed in a dichroism experiment) mainly parallel to the *c* axis. In contrast, this ferromagnetic components are confined to the *ab* plane below  $T_1$ .

In the recent XRMS experiment on GdFe<sub>4</sub>Al<sub>8</sub> at ID 20, we intended to observe the Fe moment modulation directly by tuning the x-ray energy to the Fe *K* edge. However, since we found only very weak traces of satellite reflections with a counting time of ~10 min. per single data point, we deemed extensive temperature-dependent measurements not feasible in the allotted time. Therefore, the experiment was refocused on the detailed study of the low *T* behavior using the Gd  $L_3$  resonance (7.246 keV). The experiment were performed in the cryomagnet to prevent interfering oscillations of the very small sample in the incoming beam caused by the initially used displex cryostat. Accordingly, the  $\pi$ - $\sigma$ ' geometry were realized using the MgO (222) analyzer very close to perfect with a scattering angle of 89.5 deg for the Gd  $L_3$  resonance.



Fig. 1: XRMS diffraction pattern of GdFe<sub>4</sub>Al<sub>8</sub> around the charge (440) reflection for selected temperatures.

The left panel of Fig. 1 shows XRMS diffraction pattern for selected temperatures below  $T_2$ . At T = 26.5 K, a magnetic reflection was found at (4.11 4.11 0), a satellite of the (440) charge reflection and corresponding to an incommensurate propagation vector ( $\tau\tau$ 0) with  $\tau = 0.11$ . Its intensity of ~70 cts./10 sec. is typical for the antiferromagnetic satellite reflections observed for the high *T* phase between  $T_2$  and  $T_N$ . We interpret these signals as induced by the Fe moment order. This reflection can be detected in the complete temperature range between 26.5 K and 17 K. The value for  $\tau$  increases with decreasing temperature from 0.11 to 0.128. Below 17 K the intensity rapidly increases to ~480 cts./10 sec. at 14 K. This much stronger signal can be interpreted as the manifestation of an antiferromagnetic modulation of the Gd 4*f* moments. The transition temperature to the low *T* phase is in suitable agreement with that found in magnetisation measurements on this sample.

In the narrow temperature range between 23.5 K and 26 K, we found a second set of antiferromagnetic satellite reflections in addition to the persisting reflections from higher *T* phase. Since they are similarly intense than the satellite reflections of the low *T* phase we relate them also to the Gd 4*f* moments. This interpretation is supported by specific heat data [2] indicating the onset of the Gd 4*f* moment order in this temperature range. As obvious from the right panel of Fig. 1, these magnetic satellite reflections are strongly temperature dependent. With decreasing temperature the value of  $\tau_2$  in ( $\tau_2 \tau_2 0$ ) decreases from 0.09 at 26 K to 0.04 at 23.5 K, respectively. Concomitantly, the width of the reflections increases strongly indicating a shortening in the correlation length of the antiferromagnetic order. In general, the reflections corresponding to ( $\tau_2 \tau_2 0$ ) are much broader than the ( $\tau \tau 0$ ) reflections (e. g. compare the reflections at (4.072 4.072 0) and at (4.11 4.11 0) for T = 25 K in the left panel of Fig. 1). This can be interpreted as precursor of a transition into a ferromagnetic state ( $\tau_2 = 0$ ) of the Gd 4*f* moments and as the microscopic fingerprint of the phase ITP between  $T_1$  and  $T_2$  in zero field (see Ref. [2]). Such precursor effects can also explain, why the phase ITP is strongly hysteretic in both temperature and magnetic field, and, therefore, very sensitive to variations in stoichiometry and to crystallographic imperfections [2].

To conclude definitely about the nature of this additional antiferromagnetic modulation and its relation to the two magnetic subsystems further experiments are necessary, e. g. by comparing the signals at different Gd (and probably Fe) resonances or by applying a magnetic field.

[2] M. Angst et al., PRB 72, 174407 (2005).

<sup>[1]</sup> S. Langridge et al., PRL 82, 2187 (1999); see also J.A. Paixão et al., PRB 61, 6176 (2000).