

## Magnetic Behavior of Chromium-Substituted $\text{Ho}_2\text{Fe}_{17}$ Alloys: A Temperature-Based Study

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### ABSTRACT

$\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  ( $x = 0.5, 1, 2$ ) is a ferrimagnetic system with  $T_c$  lying in the temperature range  $T = 394 - 415$  K. The magnetization of the compounds at different values of  $H$  (0.05 T – 7T) in the temperature range  $T = 4 - 380$  K have been reported. The overall behavior has been explained by the temperature and field dependent magnitude of the rare earth moment in addition to the Fe-sublattice. The lower temperature ( $<< T_c$ ) behavior is more complex and needs further studies for a clear picture.

**KEYWORDS:** Permanent magnetic material, Intermetallic compounds, Ferrimagnetism

### I. INTRODUCTION

The early research work in the  $\text{R}_2\text{Fe}_{17}$  (R-rare earth) compounds mainly encompassed on the utility of this compound as a permanent magnetic material (PMM). A PMM should possess high Curie temperature ( $T_c$ ), high saturation magnetization ( $M_s$ ) and easy-axis anisotropy. The commonly used PMM are  $\text{R}_2\text{Fe}_{14}\text{B}$  (R = Nd),  $\text{SmCo}_5$  etc. Compared to these materials,  $\text{R}_2\text{Fe}_{17}$  series of compounds contain low rare earth concentration, and hence would be economically cheaper. However, the main hindrance in using  $\text{R}_2\text{Fe}_{17}$  compounds as PMM comes from their comparatively lower  $T_c$  (although close to room temperature) [1,2] and their easy-plane anisotropy.[2] The early research work mainly focused on to increase the  $T_c$  and change the anisotropic direction from easy plane to easy axis of these compounds, by substituting other elements (magnetic: Cr, Mn, non-magnetic: Ga, Al, Si) on the Fe site as well as inserting elements like C, B, H and N in the interstices.[2,3,4,5,6,7] For the substituted compounds,  $\text{Gd}_2\text{Fe}_{16}\text{Cr}$  shows the highest  $T_c \sim 575$  K,[3] whereas the nitrogen inserted compound  $\text{Gd}_2\text{Fe}_{17}\text{C}_y\text{N}_x$  shows the maximum  $T_c \sim 764$  K.[4] However, uni-axial anisotropy has only been observed in C/N filled  $\text{Sm}_2\text{Fe}_{17}$  with a lower value of  $M_s$ .[5] So, the prospect of  $\text{R}_2\text{Fe}_{17}$  compound and their derivatives appear to be bleak as a PMM.

However, the interest in the  $\text{R}_2\text{Fe}_{17}$  compounds/its derivatives did not decay with time, rather it increased due to observation of interesting phenomena like spin reorientation, first-order magnetic phase transitions, magnetocaloric effect and negative thermal expansion (NTE) below  $T_c$ . [6,7,1,3,9,10,11,12,13] Such observations indicated the richness of the system from the fundamental as well as application point of view. The most interesting contribution of the recent study is the discovery of  $\text{Ho}_2\text{Fe}_{16}\text{Cr}$ , performing as a single component ZTE material in the temperature range  $T = 13 - 330$  K. [10]

$\text{R}_2\text{Fe}_{17}$  compounds show various types of magnetic ordering. Compounds containing lighter rare earth atoms show a collinear ferromagnetism, and those having heavier rare earth atoms show mainly ferrimagnetism.[1]  $\text{Ho}_2\text{Fe}_{17}$  compound is a ferrimagnet with  $T_c = 326$  K.[10] The Cr- substitution increases the  $T_c$  for lower concentration, and then it decreases for higher concentration of Cr. Table 1 shows the ordering temperature of the three compounds, already reported.[10,11] In this article we want to study the magnetic field ( $H$ ) and temperature ( $T$ ) dependent behavior of magnetization( $M$ ) of  $\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  ( $x = 0.5, 1, 2$ ).

**Table 1 Curie temperature of the compounds  $\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  ( $x = 0.5, 1, 2$ )**

Sample name	$T_c$ (in K)
$\text{Ho}_2\text{Fe}_{16.5}\text{Cr}_{0.5}$	394
$\text{Ho}_2\text{Fe}_{16}\text{Cr}$	415
$\text{Ho}_2\text{Fe}_{15}\text{Cr}_2$	402

## II. MATERIALS AND METHODS

$\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  ( $x = 0.5, 1, 2$ ) compounds were prepared in arc-furnace. The details of the procedure were described elsewhere.[10,11] The magnetization was measured in a SQUID VSM (M/S Quantum Design, Inc., USA) from 4–380 K.

## III. RESULTS AND DISCUSSION

The  $\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  ( $x = 0.5, 1, 2$ ) samples used here are taken from the same batch used in reference [10]. The samples are in single phase having hexagonal  $\text{Th}_2\text{Ni}_{17}$  structure with space group:  $\text{P}6_3/\text{mmc}$  (#194). The crystal structure of  $\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  is shown in figure 1. Here, Fe/Cr atoms occupy four atomic positions 4f, 6g, 12j and 12k, whereas Ho occupies 2a and 2b atomic positions.

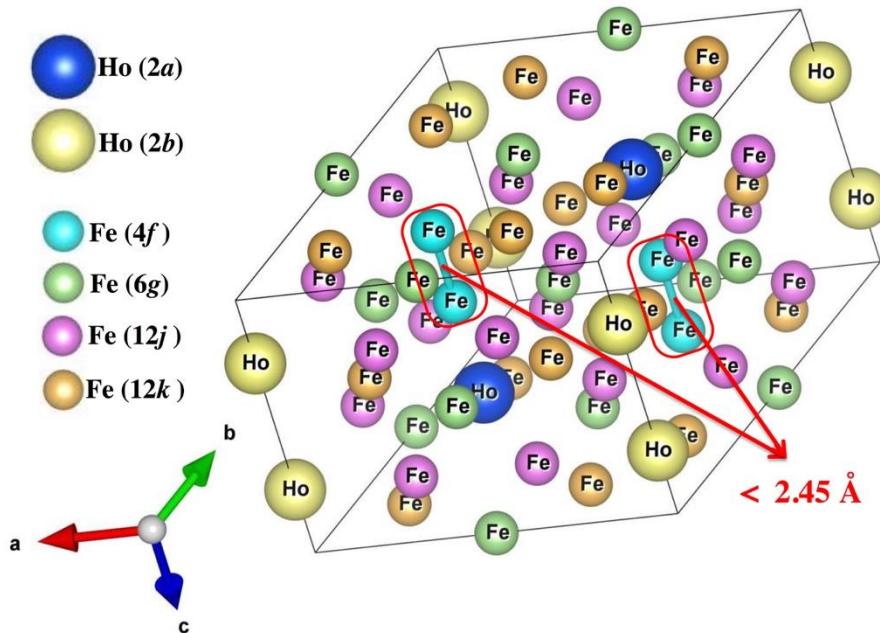


Figure 1. Crystal Structure of  $\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$

Figure 2, 3 and 4 show the behaviour of  $M$  as a function of  $T$  for the compounds  $\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  ( $x = 0.5, 1, 2$ ) at different  $H$ , measured under both zero field cooled protocol (ZFC) and field cooled (FC) protocol. With the lowering of temperature,  $M$  ( $T$ ) increases suddenly near the ferrimagnetic transition temperature  $T_C$ . The magnetism of  $\text{R}_2\text{Fe}_{17}$  compound can be explained with the help of two sublattice model, namely, rare earth sublattice and Fe sublattice. For the ferromagnetic system spins of both the sublattice orders in the same direction, whereas it is opposite for the ferrimagnetic system.[8,10] Above  $T_C$ , in  $\text{Ho}_2\text{Fe}_{17}$ , the Fe atoms at the 4f sites are coupled antiferromagnetically, while the rest of the atoms (6g, 12j and 12k) are coupled ferromagnetically.[8] Below  $T_C$ , the 4f-4f bond length increases resulting into a ferromagnetic interaction.[10]. The rare earth sublattice is coupled anti-parallel to the Fe- sublattice. The effect of substitution of magnetically weaker atom Cr at the Fe site has been discussed earlier.[10,11]

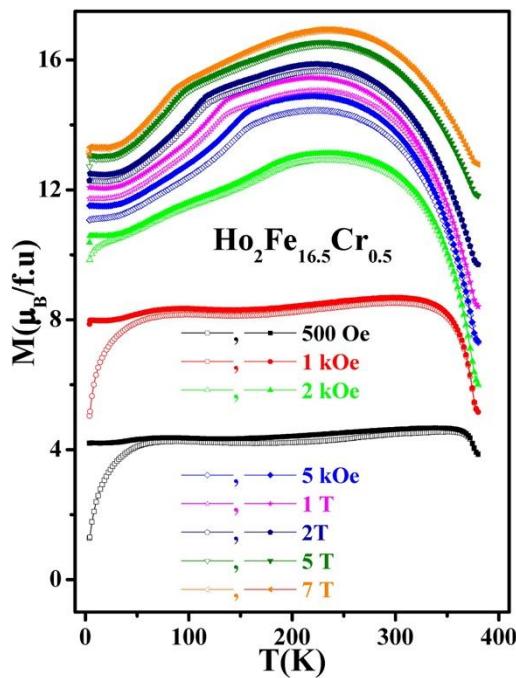


Figure 2. Magnetization ( $M$ ) of  $\text{Ho}_2\text{Fe}_{16.5}\text{Cr}_{0.5}$  as a function of temperature ( $T$ ) at different magnetic fields ( $H = 0.05 \text{ T} - 7 \text{ T}$ )

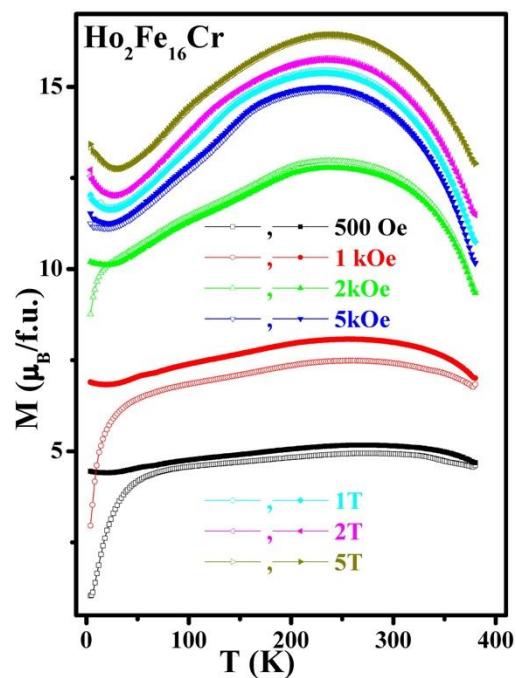


Figure 3. Magnetization ( $M$ ) of  $\text{Ho}_2\text{Fe}_{16}\text{Cr}$  as a function of temperature ( $T$ ) at different magnetic fields ( $H = 0.05 \text{ T} - 5 \text{ T}$ )

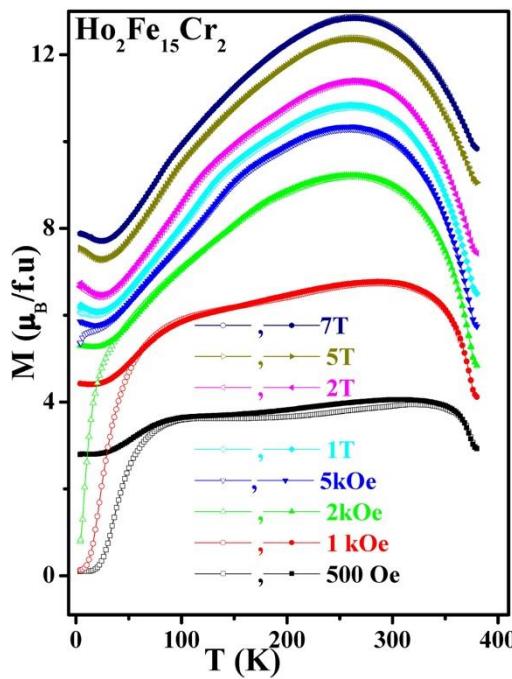


Figure 4. Magnetization ( $M$ ) of  $\text{Ho}_2\text{Fe}_{15}\text{Cr}_2$  as a function of temperature ( $T$ ) at different magnetic fields ( $H = 0.05 \text{ T} - 7 \text{ T}$ )

The salient feature of the  $M(T)$  curve at different  $H$  values are the sudden rise in the value of  $M(T)$  near  $T_C$ . This rise may be associated with ferromagnetic transition of the Fe-sublattice. At a lower value of  $H$ , ZFC and FC curves starts decreasing at temperatures much lower than  $T_C$ , whereas at higher measuring field  $M(T)$  starts decreasing at temperatures just below  $T_C$ . Such lowering of  $M(T)$  may be due to either of the two reasons: (a) the competition between the rare earth and Fe sublattice, (b) thermomagnetic history effect.[6] The thermomagnetic history effect can be explained as follows. Below  $T_C$ , in the 3d sublattice ferromagnetic domains are formed. At comparatively higher temperature, the domain walls remain free and easily respond to

the magnetic field. For strongly anisotropic system, the crystalline anisotropy causes narrow domain wall with high domain wall energy per unit area. As we lower the temperature, the thermal activation cannot provide the energy for domain wall motion. This causes to decrease  $M$  with lowering  $T$ . In such cases we should observe a decrease in  $M(T)$  just below  $T_C$  even at a lower value of  $H$ . On the other hand, we observe such behaviour only at high  $H$ . The reason lies in the increasing rare earth moment at high  $H$ , and the rare earth moment is antiparallel to Fe moment.

As we increase the measuring field  $H$  more and more, the ZFC curve approaches FC curve. Moreover, the maximum shifts to the lower temperature with increasing  $H$ . Such behaviour can be compared with the cooperative freezing of the spin glass system. Such behaviour can be related to the competing interaction of the Fe-sublattice and rare earth sublattice.

An upturn in  $M(T)$  is observed at low temperatures particularly for  $x = 1, 2$  compounds. Such upturn is associated with increasing magnetic moment of rare earth atom with the lowering of temperature.

#### IV. CONCLUSION

The behaviour of magnetization of  $\text{Ho}_2\text{Fe}_{17-x}\text{Cr}_x$  ( $x = 0.5, 1, 2$ ) as a function of temperature at different values of the applied magnetic field, both under ZFC and FC conditions have been studied. The study establishes that the changing magnitude of the rare earth moment with increasing field and temperature plays a crucial role in determining the magnetic behaviour of the studied compounds, in addition to the Fe-sublattice. The cusp around  $T_C$  reminds us of the spin glass freezing phenomenon. The lower temperature behaviour ( $<<T_C$ ) of magnetization is more complex and requires farther studies.

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