Magnetoresistance studies of $Ho_{0.85}Tb_{0.15}Mn_{2-x}Fe_x$ ($x=0$, 0.25, 0.5, 1, and 2)

J. Arout Chelvane, G. Markandeyulu,* and N. Harish Kumar
Magnetism and Magnetic Materials Laboratory, Department of Physics, Indian Institute of Technology Madras, Chennai-600 036, India

R. Nirmala and S. K. Malik
Tata Institute of Fundamental Research, Mumbai-400 005, India
(Received 16 May 2005; published 15 September 2005)

Magnetoresistance ($\Delta\rho/\rho$) studies have been carried out on the $C15$-type cubic Laves phase $Ho_{0.85}Tb_{0.15}Mn_{2-x}Fe_x$ ($x=0$, 0.25, 0.5, 1, and 2) compounds up to a magnetic field of 90 kOe at temperatures 5, 30, and 100 K. The lattice parameters were found to decrease with increasing x. The Curie temperature is found to increase with increasing x from 30 to 680 K, which is attributed to the net increase in the exchange splitting of the spin-up and spin-down bands. A maximum value of $\Delta\rho/\rho$ of -27% was observed for $Ho_{0.85}Tb_{0.15}Mn_2$ around the ordering temperature. This is explained on the basis of nearest Mn-Mn critical distance. A positive magnetoresistance, increasing in a discontinuous manner with increasing field, due to the dependence of scattering of conduction electrons on large magnetoelastic interactions, was observed at 5 K in $Ho_{0.85}Tb_{0.15}Fe_2$.

DOI: 10.1103/PhysRevB.72.092406 PACS number(s): 75.50.−y, 72.15.Gd, 75.30.Gw

INTRODUCTION

The magnetic properties of the RT_2 (R: rare earth, T: transition metal) intermetallics are of considerable interest due to the presence of localized $4f$ electrons as well as itinerant $3d$ electrons.[1,2] It has been reported that, in the binary RT_2 (T =Fe, Co, and Ni) compounds, Fe has a permanent moment, Co has an induced moment due to the R moments, and Ni has no moment.[1–6] The Fermi level is situated in such a position within the common $3d$-$4s$ band (near the large slope region of the DOS curve) where the material can display varying exchange interactions depending on the nature and relative amounts of the several transition metals. The effects of substitution at the Fe site on the magnetic properties of anisotropy compensated Ho-Tb-Fe and Dy-Tb-Fe systems have been investigated extensively.[7–9] Scattering due to spin fluctuations has been reported in Co substituted anisotropy compensated compounds viz., $Dy_{0.73}Tb_{0.27}Fe_2$ and $Ho_{0.85}Tb_{0.15}Fe_2$.[7] Spin flip metamagnetism in Co substituted $Dy_{0.73}Tb_{0.27}Fe_2$ (Ref. 8) and domain wall pinning in Ni substituted $Dy_{0.73}Tb_{0.27}Fe_2$ (Ref. 9) and have been reported.

The RMn_2 compounds crystallize in the cubic $C15$-type structure for R=Gd, Tb, Dy, Ho, and Y and in the hexagonal $C14$-type structure for R=Pr, Nd, Sm, Er, Tm, Yb, Lu, and Th.[10] The magnetic ground state of Mn in RMn_2 compounds is reported to strongly depend on the Mn-Mn nearest-neighbor distance $d_{\text{Mn-Mn}}$ and an antiferromagnetic ground state has been reported for Mn moments in compounds with $d_{\text{Mn-Mn}}$ greater than a critical distance (d_c=2.66 Å).[10–16] Malik and Wallace[12] have studied magnetic behavior of $C15$-type RMn_2 compounds, and a virtual bound state picture of $3d$ electrons was suggested to explain the structural changes and collapse of the Mn moment. For the compounds with $d_{\text{Mn-Mn}}$ greater than the critical value the Mn moment evolves from a spin fluctuating state at high temperatures to a localized moment[10–19] as the temperature is lowered. The appearance of the localized moment coincides with the onset of antiferromagnetic order with a marked increase in the volume of the unit cell. In contrast, for compounds with $d_{\text{Mn-Mn}}$ less than the critical value, no such magnetovolume effect is observed. In fact, the Mn atoms do not carry any magnetic moment when $d_{\text{Mn-Mn}}$ is less than the critical value.

One of the means to modify the character of $3d$ electrons in RMn_2 compounds is by changing the nearest Mn-Mn spacing by partly replacing Mn by another atom such as Fe, Al, etc.[14–19] Such substitutions result in increase of the Mn/Fe(Al)-Mn/Fe(Al) distance to more than the critical value where the character of the $3d$ electrons changes from itinerant to localized. Recently the effects of Mn substitution on the magnetic properties of anisotropy compensated $Ho_{0.85}Tb_{0.15}Fe_2$ and $Dy_{0.73}Tb_{0.27}Fe_2$ have been investigated and spin fluctuations have been reported near the Curie temperature (T_C)[18,19] when the $d_{\text{Mn/Fe-Mn/Fe}}$ distance is close to the critical distance. These fluctuations when suppressed by the application of an external magnetic field can lead to the observation of large magnetoresistance in these compounds, caused by the spin polarization of $3d$ electrons and subsequent s-d transitions. We have, therefore, investigated the magnetic field dependence of electrical resistivity of $Ho_{0.85}Tb_{0.15}Mn_{2-x}Fe_x$ and the results are presented in this paper.

EXPERIMENTAL DETAILS

The compounds were prepared by arc melting the constituent elements in an arc furnace under argon atmosphere and the ingots were homogenized in vacuum at 1170 K for a week and were then furnace cooled.[18] Structural characterization was carried out by powder x-ray diffraction studies employing Fe-K_α radiation.[18] Magnetization measurements were carried out using a SQUID magnetometer (MPMS XL, Quantum Design) in the temperature range 5–300 K and a PAR 155 vibrating sample magnetometer in the temperature range 300–700 K. Electrical resistance as a function of magnetic field was measured by a conventional four probe

TABLE I. The lattice constants (Mn,Fe)-(Mn,Fe) distances and Curie temperature in $Ho_{0.85}Tb_{0.15}(Mn_{2-x}Fe_x)$ alloys.

x	a (±0.001 Å)	$d_{(Mn\text{-}Fe)\text{-}(Mn\text{-}Fe)}$ (±0.001 Å)	T_C (K)
0	7.547	2.668	30
0.25	7.527	2.661	48
0.5	7.499	2.650	130
1	7.444	2.632	249
2	7.310	2.584	620

method at selected temperatures using a physical property measurement system (PPMS, Quantum Design), up to a magnetic field of 90 kOe.

RESULTS AND DISCUSSIONS

All the compounds $Ho_{0.85}Tb_{0.15}Mn_{2-x}Fe_x$ with $x=0$, 0.25, 0.5, 1, and 2 were found to form in single phase with $C15$ cubic Laves phase structure. The lattice parameters decrease with increasing Fe concentration in accordance with the relative metallic radii of Fe and Mn (Ref. 18, Table I). The nearest Mn/Fe-Mn/Fe calculated from the structure are displayed in Table I.

The Curie temperature is seen to increase from 30–680 K with decreasing Mn concentration and the values for all the compounds investigated are shown in Table I and the variation has been explained on the basis of reduction in exchange splitting of the spin-up and spin-down $3d$ bands.[18] Similar variations in T_C has been reported in $Tb(Mn_{1-x}Fe_x)_2$ (Ref. 17) and $Dy_{0.73}Tb_{0.27}Fe_{2-x}Mn_x$ (Ref. 19) systems.

The magnetoresistance is defined as $\Delta\rho/\rho=[\rho(H)-\rho(0)]/\rho(0)\times 100$, where $\rho(H)$ is the electrical resistivity in the presence of a magnetic field (H) and $\rho(0)$ is the resistivity in absence of a magnetic field.

The $\Delta\rho/\rho$ obtained for $Ho_{0.85}Tb_{0.15}Mn_2$, as a function of magnetic field, at 5, 25, 50, and 100 K are shown in Fig. 1. A magnetoresistance of about −27%, the first in any RMn_2 system, at 90 kOe is observed at a temperature of 25 K (close to 30 K, the Curie temperature). This is explained on the basis of the sensitivity of the Mn moments to the interatomic distances in RMn_2 systems. In the present case, the nearest Mn-Mn ($d_{Mn\text{-}Mn}$) distance is 2.668 Å, almost the critical $d_{Mn\text{-}Mn}$ distance. It has been reported that just above the critical distance the antiferromagnetic ordering of Mn cannot sustain the negative interaction between all the Mn moments and this causes the system to be frustrated, resulting in spin fluctuations especially at temperatures close to T_C.[14,20] Under the influence of a magnetic field, due to the alignment of the Mn moments in the direction of the field, the scattering and hence the resistance decreases. In $Ho_{0.85}Tb_{0.15}Mn_2$ the $\Delta\rho/\rho$ due to this reduction seems to be as large as −27%. The $\Delta\rho/\rho$ value is found to be around −21% at 5 K, probably due to the increased ordering of the Mn moments.

For the compound with $x=0.25$, in which the nearest (Mn/Fe)-(Mn/Fe) distance $[d_{(Mn/Fe)\text{-}(Mn/Fe)}]$ is 2.661 Å, the

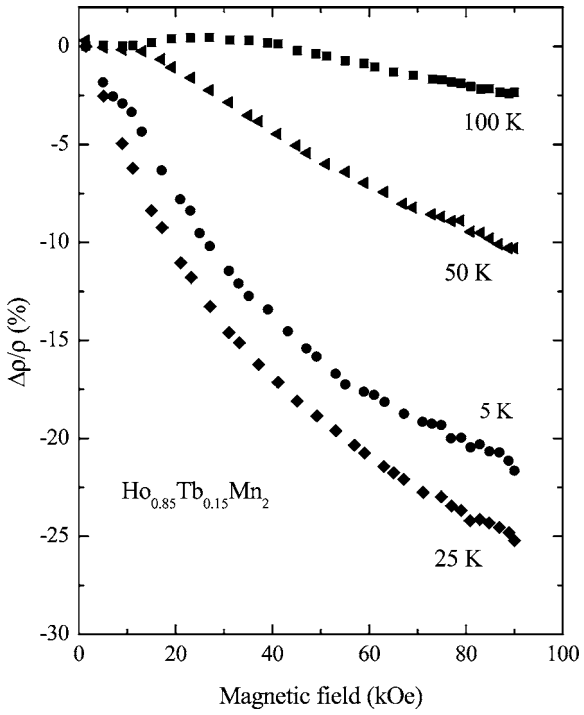

FIG. 1. Magnetoresistance of $Ho_{0.85}Tb_{0.15}Mn_2$ at 5, 25, 50, and 100 K.

$\Delta\rho/\rho$ at 30 and 5 K are found to be −10% and −5% (Fig. 2), respectively. This is in accordance with the higher Curie temperature (50 K) of this material (Table I) leading to a reduced magnitude of $\Delta\rho/\rho$ compared to that in $Ho_{0.85}Tb_{0.15}Mn_2$. With increasing iron concentration the

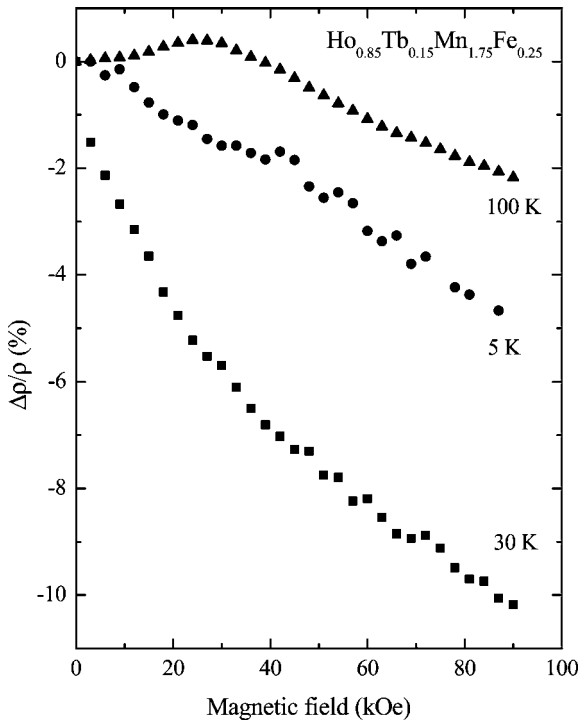

FIG. 2. Magnetoresistance of $Ho_{0.85}Tb_{0.15}Mn_{1.75}Fe_{0.25}$ (values are less than those of $Ho_{0.85}Tb_{0.15}Mn_2$).

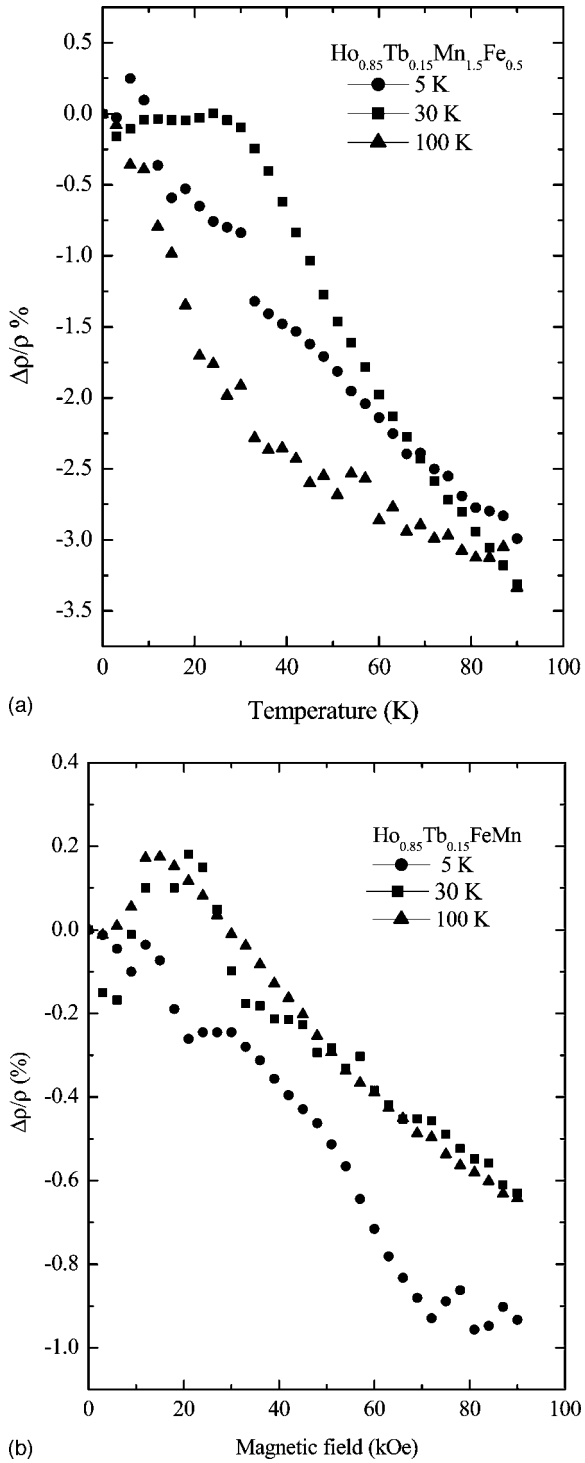

FIG. 3. (a) Magnetoresistance of $Ho_{0.85}Tb_{0.15}Mn_{1.5}Fe_{0.5}$, (b) magnetoresistance of $Ho_{0.85}Tb_{0.15}MnFe$.

magnetic moment of the 3*d* sublattice and the Curie temperature of the compound increase and thus the moments are well ordered at 5 K. Therefore, the enhanced ordering of magnetic moments should cause a reduction in the scattering compared to that in $Ho_{0.85}Tb_{0.15}Mn_2$ and this in fact is observed in compounds with $x=1.5$, 1, and 2, as shown in Figs. 3(a), 3(b), and 4.

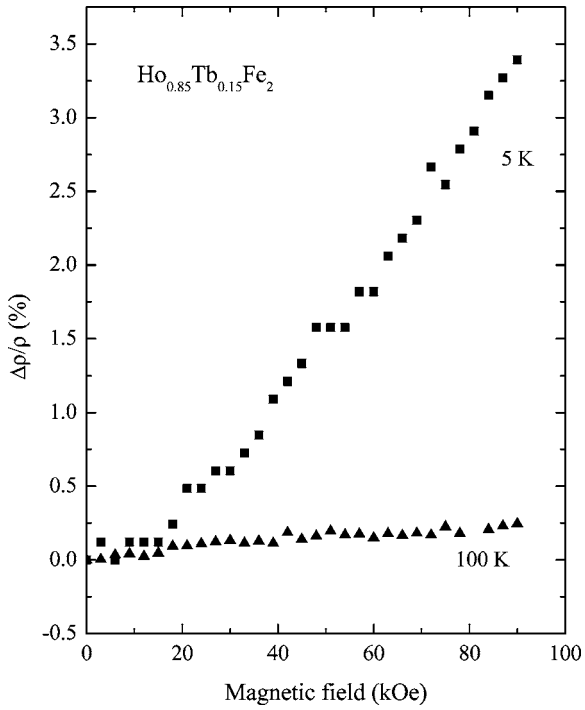

FIG. 4. Positive magnetoresistance of $Ho_{0.85}Tb_{0.15}Fe_2$ with magnetic field.

Another interesting feature is the positive sign of $\Delta\rho/\rho$ at 5 K for the compound $Ho_{0.85}Tb_{0.15}Fe_2$, one of the compounds possessing giant magnetostriction driven by large strain induced magnetoelastic interactions.[2] While at room temperature the localized anisotropies of the Ho and Tb sublattices are nearly compensated for, at low temperatures considerable increase in the localized anisotropy is further enhanced by the application of the magnetic field that results in enhanced distortion. This could cause an increase in the spin-dependent scattering and hence the resistivity.

In $Ho_{0.85}Tb_{0.15}Mn_2$, $\Delta\rho(90\ \text{kOe})/\rho$ at 50 K (in its paramagnetic state) is found to be −9% (Fig. 1). This could be due to the combined effect of paramagnetism and spin fluctuations. At 100 K, the paramagnetism dominates and hence the value of $\Delta\rho/\rho$ decreases to −2.5%. With the decrease in Mn concentration, $\Delta\rho/\rho$ is found to be very small.

The positive value of $\Delta\rho/\rho$ up to a field of 40 kOe at 100 K for $x=0$ and 0.25, where they are paramagnetic, is rather surprising and we do not understand the reason for this behavior.

SUMMARY

The effect of Fe substitution on the magnetoresistance of $Ho_{0.85}Tb_{0.15}Mn_2$ has been investigated. The compound $Ho_{0.85}Tb_{0.15}Mn_2$ is found to exhibit a maximum magnitude of $\Delta\rho/\rho$ (27%) around the ordering temperature which is attributed to the spin fluctuations. The $|\Delta\rho/\rho|$ with increasing iron concentration decreases and is attributed to reduction in spin fluctuations. $Ho_{0.85}Tb_{0.15}Fe_2$ exhibits a positive magnetoresistance.

ACKNOWLEDGMENTS

One of the authors, J. Arout Chelvane, thanks the Indian Institute of Technology Madras for financial support.

*Email address: mark@physics.iitm.ac.in

[1] K. H. J. Buschow, Rep. Prog. Phys. **40**, 1179 (1977).
[2] A. E. Clark, in *Ferromagnetic Materials*, edited by E. P. Wohlfarth (North Holland, Amsterdam, 1980), p. 531.
[3] N. C. Koon, C. M. Williams, and B. N. Das, J. Magn. Magn. Mater. **100**, 173 (1991).
[4] K. H. J. Buschow, Rep. Prog. Phys. **54**, 1123 (1991).
[5] A. E. Clark and H. S. Belson, in *Magnetism and Magnetic Materials*, edited by C. D. Graham and J. J. Rhyne, AIP Conf. Proc. No. 10 (AIP, New York, 1972), p. 749.
[6] N. C. Koon, A. I. Schindler, C. M. Williams, and F. L. Carter, J. Appl. Phys. **45**, 5389 (1974).
[7] K. R. Dhilsha and K. V. S. Rama Rao, J. Appl. Phys. **68**, 259 (1990).
[8] S. Kishore, G. Markandeyulu, and K. V. S. Rama Rao, Solid State Commun. **108**, 313 (1998).
[9] M. Senthil Kumar, K. V. Reddy, and K. V. S. Rama Rao, IEEE Trans. Magn. **31**, 4160 (1995).
[10] M. Shiga, Physica C **149B**, 293 (1988).
[11] M. Shiga, J. Magn. Magn. Mater. **129**, 17 (1994).
[12] S. K. Malik and W. E. Wallace, J. Magn. Magn. Mater. **24**, 23 (1981).
[13] C. Ritter, M. R. Ibarra, and C. Marquina, J. Magn. Magn. Mater. **196-197**, 721 (1999).
[14] B. Ouladdiaf, J. Deportes, R. M. Galera, A. S. Markosyan, and J. L. Soubeyroux, Physica B **180-181**, 85 (1992).
[15] S. Labroo, N. Ali, and P. Robinson, J. Appl. Phys. **67**, 5292 (1990).
[16] M. T. F. Telling, C. Ritter, and R. Cywinski, Physica B **276-278**, 740 (2000).
[17] W. Zhang, E. M. Levin, and K. A. Gschneidner, Jr., J. Magn. Magn. Mater. **250**, 170 (2002).
[18] J. Arout Chelvane, S. Kasiviswanathan, and G. Markandeyulu, J. Magn. Magn. Mater. **277**, 175 (2004).
[19] J. Arout Chelvane and G. Markandeyulu, J. Magn. Magn. Mater. **294**, 298 (2005).
[20] K. Krop, Physica B **319**, 9 (2002).