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Magnetic and electrical studies on $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$ system

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Cubic Laves phase alloys of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$ $(0<x<2)$ were prepared and the lattice parameters, evaluated as a function of x at room temperature, show deviation from Vegard's law. This was attributed to the presence of a magnetovolume effect and charge transfer. Magnetization measurements were performed in the temperature range 77–700 K. The results suggest that the system undergoes a gradual transition from ferri- to ferromagnetic with increasing x. This has been explained based on the $3d$ band model. Furthermore, a monotonic decrease in Curie temperature with x was observed. Electrical resistivity studies were carried out in the temperature range 16–700 K. An anomaly was found in the vicinity of the Curie temperature for $x=2$ which has been attributed to the existence of the long-range nature of the critical fluctuations due to short-range spin ordering.

I. INTRODUCTION

RE-TM (where RE denotes rare earth and TM denotes transition metal) alloys find extensive applications as magnetostrictive materials, permanent magnets, etc.[1–3] Studies on these alloys are essential in understanding the nature of interactions which play a crucial role in determining various magnetic properties. Among these, $RETM_2$ systems with Fe, Co, and Ni form excellent tools for the study of d magnetism. A greater understanding of the nature of the magnetic interactions can be obtained by studying the changes in magnetic properties occurring when one TM is gradually replaced by another TM without change in the crystal structure. The $3d$ electrons in these systems are itinerant in nature and therefore the formation of a $3d$ band takes place.[4,5] By changing the composition, the Fermi level can be varied and the position of this level determines the magnetic moment on the TM site. Neutron-diffraction results showed that, in $RETM_2$ systems, Fe carries a magnetic moment, Ni does not, whereas Co has an induced moment.[6–8] These studies also confirmed that $REFe_2$ and $RECo_2$ are ferrimagnetic and $RENi_2$ is ferromagnetic. Electrical resistivity measurements near the Curie temperature T_C in these systems are extremely sensitive to the nature of the magnetic interactions.

Among the $RETM_2$, $REFe_2$ systems exhibit magnetostriction an order of magnitude larger than any other material known at present. These systems are found to have large magnetic anisotropy and, therefore, high magnetic fields are required to saturate the materials. Clark and Belson[9] and Koon *et al.*[10] have found that the anisotropy-compensated $Dy_{0.73}Tb_{0.27}Fe_2$ and $Ho_{0.85}Tb_{0.15}Fe_2$ alloys possess low magnetic anisotropy still retaining large magnetostriction which can be used for practical applications.[1,2] In our laboratory, studies on the substitution at the Fe site and interstitial hydrogen have been carried out extensively. Investigations on the effect of Co in these two alloys on the magnetic, magnetomechanical, and electrical properties gave interesting results.[11–13] The effect of interstitial hydrogen on the magnetic and electrical properties in these systems also has been investigated.[14–16] In this article we present the results of the effect of Ni in $Ho_{0.85}Tb_{0.15}Fe_2$ on the lattice parameter, magnetization, and electrical resistivity.

II. EXPERIMENT

The samples were prepared by melting the constituent elements, Ho and Tb of purity 99.9% and Fe and Ni of purity 99.99%, in an arc furnace under argon atmosphere. Meltings were carried out several times and each time the melted ingots were turned upside down to ensure homogeneity. Weight losses after final melting were less than 0.6% for all the samples prepared. The samples were then annealed in evacuated quartz tubes at 1170 K for 10 days.

Powder-x-ray diffractograms were taken using Co $K\alpha$ radiation and the lattice parameters were evaluated by least-squares refinement. Magnetization measurements were performed using a PAR 155 vibrating sample magnetometer up to an applied field of 10 kOe. Temperature variation from 77 K to room temperature (RT) was carried out by the continuous-flow method using liquid nitrogen and an oven assembly was used to achieve high temperatures. Temperatures were maintained within ± 1 K.

Electrical resistivity ρ measurements were carried out using a four-probe technique.[17] Samples for the measurements were initially cut from the ingots using a diamond cutter and then brought to the shape of disks of approximately 6 mm diameter and 1 mm thickness after polishing. Then the samples were reannealed at 1000 K for 24 h. For carrying out measurements at low temperatures, the four contacts were made on the flat surface of the sample using silver paste. For high-temperature experiments, pressure contacts using platinum tips were employed. A constant-current source delivering 90 mA and a Keithley 181 nanovoltmeter were employed for measuring electrical resistivity.

Measurements were carried out down to 16 K using a Leybold closed-cycle helium refrigerator. Experiments

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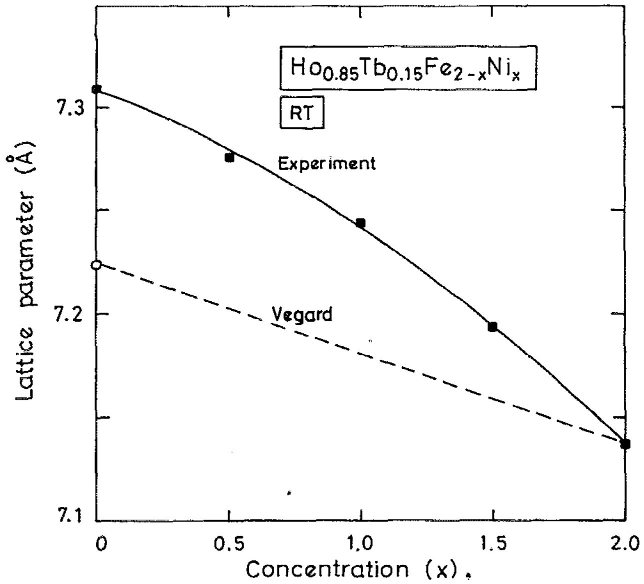

FIG. 1. Variation of lattice parameter with x at RT of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$. The "Vegard" lattice parameter for $x=0$ is calculated from the data taken from Ref. 18.

were conducted by keeping the sample in a helium exchange gas chamber. A silicon diode sensor was used for measuring the sample temperature and another diode fixed to the cold-head second stage was utilized by the Leybold LTC 60 low-temperature controller for the purpose of controlling the temperature. Temperatures were maintained within ±0.1 K. High-temperature measurements were also carried out in a helium exchange gas atmosphere. A Chromel-Alumel thermocouple was used as the temperature sensor and a resistance furnace for attaining high temperatures. Temperatures were maintained within ±1 K.

III. RESULTS AND DISCUSSION

Samples of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$ with $x=0$, 0.5, 1.0, 1.5, and 2.0 were prepared and the formation of single C15-type cubic Laves phase was confirmed from the x-ray diffractograms. Lattice parameters evaluated at RT from the diffractograms decrease with increasing Ni concentration as shown in Fig. 1. The ionic radius of Ni is smaller than Fe. If the size effect alone is taken into account, the variation of lattice parameter should follow Vegard's law. However, a nonlinear behavior is observed in the present system. Similar variations of lattice parameters were reported earlier on $RE(Fe_{1-x}Co_x)_2$ and $Y(Fe_{1-x}Co_x)_2$.[11,12,18]

The deviation from linearity can be due to two different mechanisms occurring in the present system: They are the magnetovolume effect and the charge transfer.

In 3*d* transition metals and their alloys, the volume in a magnetically ordered state is larger than that in a paramagnetic state. This is due to the presence of the magnetovolume effect (MVE), which plays a crucial role in determining the lattice parameters of the samples in the magnetically ordered state.[19] In $RETM_2$ systems with Fe, Co, and Ni, band formation takes place due to the itinerant nature of the 3*d* electrons. The splitting of bands in the magnetically ordered state is responsible for this volume expansion, whereas, the bands are unsplit in the paramagnetic state. In the present system, the samples with $x=0$, 0.5, and 1.0 are ferrimagnetic at RT and therefore the MVE is expected to be present.

On the other hand, it is well known that there is a charge transfer taking place from RE to the 3*d* band in these $RETM_2$ systems.[4,5] The variation of lattice parameter with x can also deviate from linear behavior, if the charge transfer taking place is not uniform throughout the Ni concentration.

The role of MVE in $Y(Fe_{1-x}Co_x)_2$ was observed by Eriksson *et al.*[18] They have estimated the hypothetical paramagnetic lattice parameters, as predicted by Vegard's law, for $Y(Fe_{1-x}Co_x)_2$ as a function of x. The experimentally observed data lie well above the estimated values in the Fe-rich region and the difference becomes smaller with increasing x and a good agreement is reached for $x=2$, being paramagnetic. A volume expansion of 3.5% of that of a hypothetical paramagnetic state for YFe_2, zero for YCo_2, and intermediate for other concentrations are obtained from the calculated *d* pressure due to the band magnetism of the 3*d* electrons. A reasonably good fit between experiment and theory has been obtained indicative of the dominant role played by the MVE.

Due to the strong similarity of Y to the rare earths, the 3*d* band in $REFe_2$ is the same as that in YFe_2 and the effect of the additional RE-3*d* interaction is only to split the up and down spin bands more.[20] Therefore, the MVE in $REFe_2$ will be equal to or greater than that in YFe_2. Hence, it can be assumed that the same percentage of increase in the lattice parameter of YFe_2 occurs in the present system as well. Thus, the hypothetical paramagnetic volume for $Ho_{0.85}Tb_{0.15}Fe_2$ is represented in Fig. 1 by an open circle. The dashed line joining $x=0$ and 2 is the variation of lattice parameter as predicted by Vegard's law. It can be seen that, for $x=1.5$, the lattice parameter deviates from Vegard's law, even though the sample is paramagnetic at RT. This suggests that the effect of nonuniform charge transfer is also possible in the present system.

Magnetization as a function of applied field at RT is shown in Fig. 2. Temperature variation of magnetization at an applied field of 10 kOe from 77 to 700 K for various Ni concentrations is as shown in Fig. 3. The inset shows the magnetization at an applied field of 10 kOe as a function of x at RT and 77 K.

The nature of variation of the magnetic moment of the TM sublattice was explained based on a band model due to the itinerant nature of the 3*d* electrons.[4,5] This model considers the 3*d* band as rigid and that charge transfer takes place from the RE to the common 3*d* band. From the earlier investigations, it was concluded that the Fermi level in the $REFe_2$ system lies in the vicinity of the maximum of the density of states (DOS) at the spin-up sub band and at the minimum of the spin-down sub band, and therefore Fe has an intrinsic moment. In $RECo_2$, the spin-up sub band is completely filled and the spin-down sub band is partially filled, and a large exchange field exists due to the RE,

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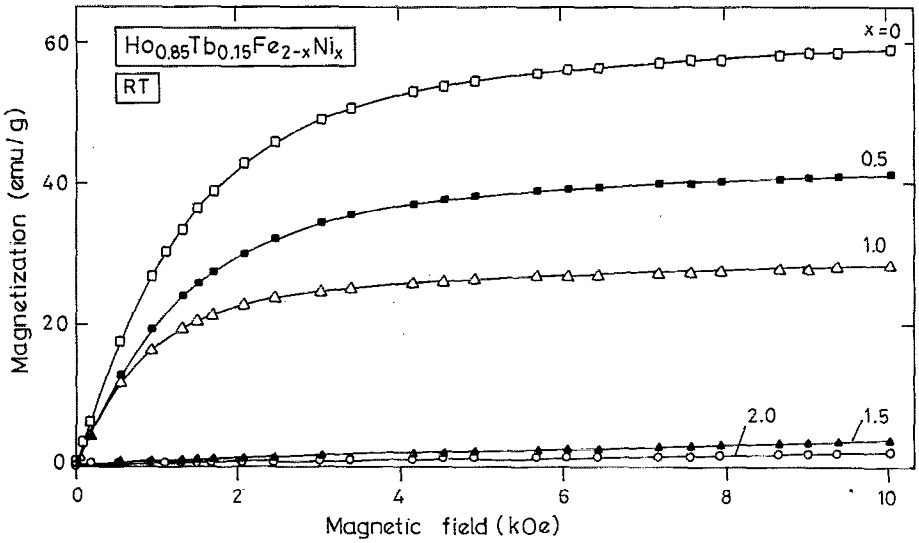

FIG. 2. Variation of magnetization as a function of applied magnetic field for various values of x at RT of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$.

resulting in an induced magnetic moment on the Co. In the $RENi_2$ system, the Fermi level is situated at the top of the $3d$ band where the DOS for both up and down spins is very small and so the band is completely filled. Consequently, Ni does not possess a magnetic moment. Neutron-diffraction results and band-structure calculations carried out on $REFe_2$, $RECo_2$, and $RENi_2$ confirmed the band model.[6,7,8,20]

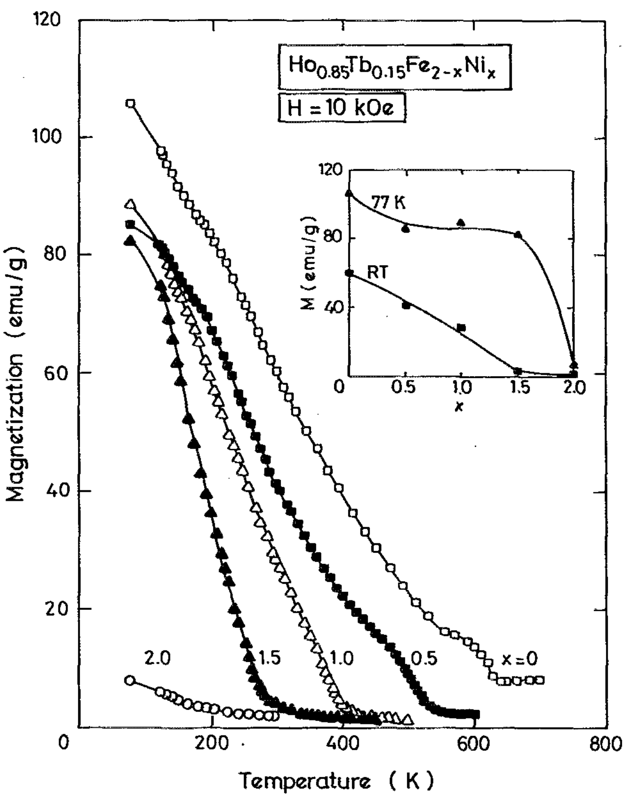

FIG. 3. Temperature variation of magnetization at an applied field of 10 kOe for various x of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$. The inset shows the magnetization at an applied field of 10 kOe as a function of x at RT and 77 K.

When Fe is gradually replaced by Ni, progressive filling up of the $3d$ band takes place which will lead to a decrease in the magnetic moment of the TM sublattice. Since $REFe_2$ is ferrimagnetic and the RE moment is higher than the Fe moment, the reduction in the moment of TM sublattice will cause an increase in the net moment per formula unit with x. It can be seen from Fig. 3 that there is a crossover occurring for $x=0.5$ and 1.0 and at 77 K the magnetization for the samples with $x=0.5$–1.5 is almost the same which is a situation different from that at RT. The increasing steepness of the curves, for $x=0$–1.5, below T_C with increasing x, suggests that the crossover for higher x will occur at lower temperatures. Similar observations were reported in $RE(Fe_{1-x}Co_x)_2$ systems.[11,12]

Figure 4 shows the T_C values determined from magnetization measurements. The value of T_C for $x=2$ was obtained from the weighted average of the T_C values of $HoNi_2$ and $TbNi_2$ from the data of Buschow.[4] As seen from the figure, the gradual replacement of Fe by Ni leads to a progressive decrease in T_C with x. It is evident that the T_C values of $RENi_2$ systems are very small due to the weak RE-RE interactions.[5] Mössbauer results showed that even the presence of 2 wt % of enriched ^{57}Fe in $GdNi_2$ raised the ordering temperature by as much as 40 K, which implies that the effect of Fe on the exchange interactions is very strong.[21] The rapid fall in T_C with x also confirms that the TM-TM and RE-TM interactions are decreasing at a faster rate. The smoothness of the T_C vs x curve indicates that the transition from ferri- to ferromagnetic state, with increasing x, is gradual. A similar decrease in T_C was observed in $Dy(Fe_{1-x}Ni_x)_2$ and $Tb_{0.3}Dy_{0.7}(Fe_{1-x}Ni_x)_{1.9}$ systems.[22,23] In the case of $RE(Fe_{1-x}Co_x)_2$ systems, an initial increase in T_C was observed up to $x=0.25$ and then it decreases.[11,12,24] Due to the existence of induced moment

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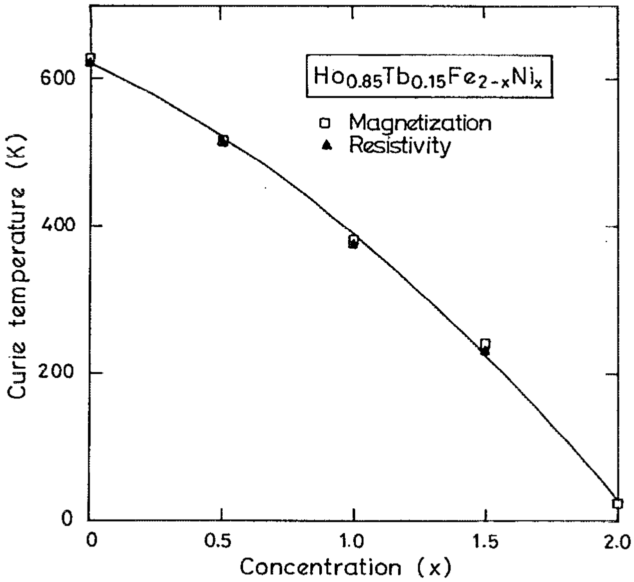

FIG. 4. Curie temperature as a function of x obtained from magnetization and resistivity measurements of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$. The value for $x=2$ is taken as the weighted average of the T_C values of $HoNi_2$ and $TbNi_2$ from Ref. 4.

on Co, the fall in T_C with x is rather slow when compared with the present system.

The temperature variation of ρ is shown in Fig. 5. The inset shows the data for $x=2$ in the temperature range 16–50 K. The increase in ρ with increasing temperature for all x suggests that all the samples exhibit metallic behavior. T_C values obtained from the resistivity measurements are also shown in Fig. 4 along with those from the magnetization data. The values of T_C obtained from both measurements are in good agreement.

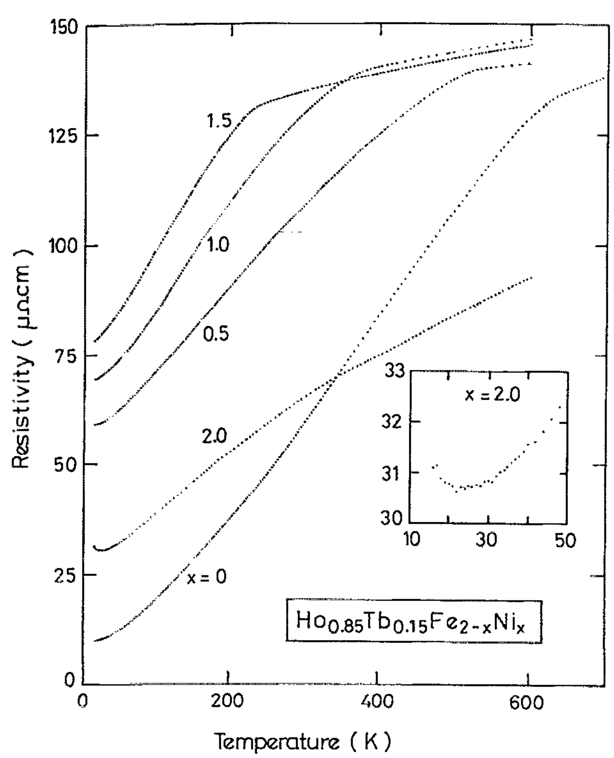

FIG. 5. Temperature variation of resistivity for various values of x of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$. The inset shows the anomaly observed for $x=2$ in the vicinity of T_C.

An anomaly in the form of a minimum is observed near T_C ($\sim$23 K) for $x=2$. Kawatra *et al.* have observed similar results in the case of $GdNi_2$.[25,26] In their ρ-T plot, ρ increases with increasing temperature up to T_C, a change of slope occurs at T_C with a flat region extended to approximately 10 K above T_C, and a linear increase when the temperature is raised further. A sharp discontinuity at T_C in the $d\rho/dT$ vs T curve was observed. This behavior was attributed to the role of the long-range nature of the critical fluctuations due to the short-range spin ordering in the framework of the molecular-field theory just above T_C.[27] The model considers the effect of s-f exchange interaction between the conduction and the localized f electrons on the electrical resistivity near the magnetic transition. This interaction is responsible for the ordering of the $4f$ moments of the rare earth and the fluctuations occurring when the sample undergoes a transition from para- to ferromagnetic state are responsible for the anomaly observed. They were able to give a good theoretical fit to the experimental data above T_C which suggested the essential correctness of the molecular-field treatment of these fluctuations. According to the theoretical treatment, these fluctuations should lead to a positive slope below T_C and a negative slope just above T_C, in the ρ-T curve. Since the T_C is around 75 K for $GdNi_2$, the existence of electron-phonon interaction smooths the divergence and the negative slope could not be observed. In our measurements the observation of a minimum was possible because of the relatively low T_C where the electron-phonon interaction is expected to be very small. The disappearance of critical fluctuations due to the development of complete ordering will lead to a decrease in ρ with decreasing temperature. This was not observed due to the lack of data below 16 K. For $x<2$, the Fe-Fe interaction is stronger and, therefore, the large molecular field produced by the exchange ordering of the Fe spins is responsible for the rare-earth spin ordering which results in the high Curie temperatures as discussed earlier.[6] Thus, the magnitude of the s-f interaction is negligible in these cases.

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