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Effect of Ni substitution on the magnetic and electrical properties of $Dy_{0.73}Tb_{0.27}Fe_2$

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Alloys of $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$ $[0 \leqslant x \leqslant 2]$ were prepared and the lattice parameters obtained at room temperature showed a deviation from Vegard's law owing to the presence of magnetovolume effect. Magnetization studies were carried out up to an applied field of 10 kOe in the temperature range 4.2–750 K. Magnetization data at 4.2 K indicate the effect of domain wall pinning. Temperature dependence of magnetization curves showed maxima at low temperatures as a consequence of this pinning effect. Electrical resistivity studies were carried out in the temperature range 16–750 K. A minimum observed for $x=2$ in the vicinity of its Curie temperature is attributed to the presence of critical fluctuations due to short-range spin ordering of the rare earths. All the samples, except $x=2$, showed a T^2 dependence at low temperatures and this has been attributed to electron-spin wave scattering mechanism. © *1995 American Institute of Physics.*

I. INTRODUCTION

$REFe_2$ (R—rare-earth) alloys, particularly $Dy_{0.73}Tb_{0.27}Fe_2$ and $Ho_{0.85}Tb_{0.15}Fe_2$, exhibit large magnetostriction and therefore, find extensive applications as magnetostrictive device materials.[1,2] The magnetic properties of these $REFe_2$ alloys are of great interest due to the presence of localized $4f$ electrons and itinerant $3d$ electrons. The magnetic anisotropy of rare earths is due to the effect of crystalline electric fields acting on the highly anisotropic $4f$ electron distribution. The $3d$ electrons show very small anisotropy due to the quenching of orbital angular momentum. Therefore, the net anisotropy of $REFe_2$ systems arises mainly due to the RE ions. In contrast, the $3d$ electrons in these alloys form narrow bands and the Fermi level is situated in a position where it can display different kinds of magnetic properties when Fe is replaced by another transition metal (TM).[3,4] It has been reported that, in $RETM_2$ (TM =Fe, Co and Ni) systems, Fe has a permanent moment, Co has an induced moment due to the RE moments, and Ni has no moment. Thus, $REFe_2$ and $RECo_2$ are ferrimagnets whereas $RENi_2$ is a ferromagnet. The Curie temperatures (T_C) of these systems are determined primarily by the TM-TM interactions. Therefore, the changes in magnetic behavior occurring when Fe is replaced by Ni, can furnish a greater understanding of the nature of magnetic interactions present in these systems.

In our laboratory, as part of the program of understanding the effect of substitution of Ni and Co in place of Fe on the structural, magnetic, and electrical properties of $Ho_{0.85}Tb_{0.15}Fe_2$ and $Dy_{0.73}Tb_{0.27}Fe_2$, we report here, the results obtained from the studies on the $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$ system. Earlier investigations carried out on $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$, $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$, and $Dy_{0.73}Tb_{0.27}Fe_{2-x}Co_x$ systems gave interesting results.[5–8]

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II. EXPERIMENT

The alloys were prepared by arc melting the constituent elements Dy and Tb of 99.9% purity and Fe and Ni of 99.99% purity in argon atmosphere for several times to ensure homogeneity. Weight losses after the final melting were less than 0.7% for all the samples prepared. These arc-melted ingots were annealed in evacuated quartz tubes at 1170 K for 10 days.

Lattice parameters were evaluated by least-squares refinement from powder x-ray diffractograms taken using Co $K\alpha$ radiation. Magnetization studies were performed on powder samples using a PAR 155 vibrating sample magnetometer up to an applied field of 10 kOe and in the temperature range 4.2–750 K. These measurements were carried out from 4.2–150 K using a continuous flow liquid helium cryostat and from 130 K to room temperature (RT) using a continuous flow liquid-nitrogen cryostat. A high temperature oven assembly was used to perform the experiments above RT.

Electrical resistivity (ρ) studies were carried out using four-probe technique. Samples for resistivity measurements were cut from the ingots using a diamond cutter and polished to get disks of approximately 6 mm diameter and 1 mm thickness. The samples were reannealed at 1000 K for 24 h. Silver paste contacts were used for low temperature measurements and pressure contacts using platinum tips were used for high temperature measurements. A Keithley 224 constant current source delivering 90 mA and a Keithley 181 nanovoltmeter were used for measuring electrical resistivity. Experiments from 16–300 K were carried out using a closed cycle helium refrigerator.[5,9] In the temperature range 300–750 K, a high temperature oven assembly was employed.

III. RESULTS AND DISCUSSION

Powder x-ray diffractograms taken at RT for $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$ samples [$x=0$, 0.5, 1.0, 1.5, and 2.0] revealed the formation of all samples in single phase with

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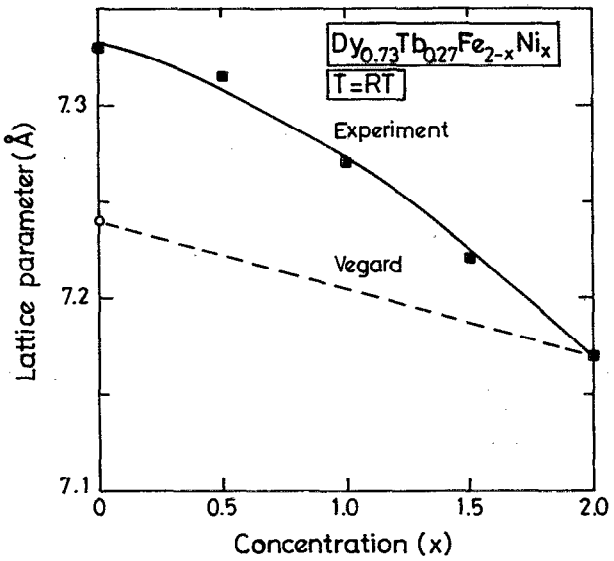

FIG. 1. Concentration dependence of lattice parameter at RT of $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$. The "Vegard" lattice parameter for $x=0$ is calculated from the data taken from Ref. 12.

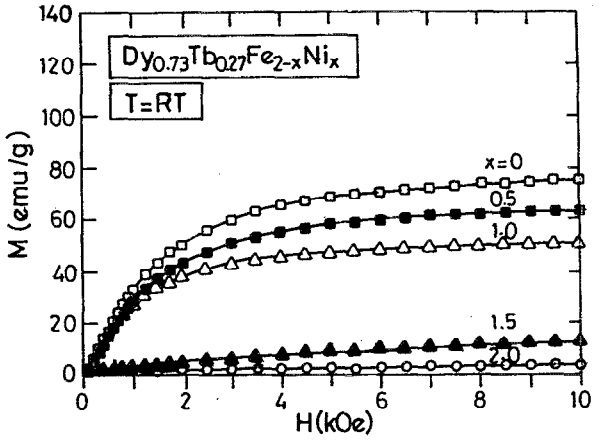

FIG. 2. Variation in magnetization with applied magnetic field for various values of x at RT.

C15-type cubic Laves phase structure. The variation in lattice parameter as a function of x exhibits deviation from linear behavior as shown in Fig. 1.

Various factors such as size factor, ionic radii, and magnetic properties have been found to influence the lattice parameters of alloys considerably.[10] The major contributions to the deviation from Vegard behavior in the present system have been attributed to the magnetovolume effect[11] (MVE) which arises due to band polarization and local moments at the $3d$ site, resulting in lattice expansion. The effect of charge transfer has influenced these deviations to a very small extent.

Eriksson *et al.*[12] have obtained a volume expansion of about 3.5% in the case of YFe_2. Since the electronic structure of YFe_2 is similar to that of $REFe_2$, the percentage of deviation of the experimental lattice parameter from the hypothetical Vegard lattice parameter for $x=0$ can be assumed to be the same as that for YFe_2.[13] However, for $x=2.0$, the Vegard lattice parameter and the experimental value are assumed to be the same and the reasons for this will be explained later. Therefore, the dashed line shown in Fig. 1 is the variation in lattice parameter as predicted by Vegard's law.

The deviation from Vegard behavior could be explained based on the unified model which includes the features of both itinerant band model and local moment model. In $RETM_2$ systems (in the present case TM=Fe and Ni), the $3d$ electrons of the transition metals are itinerant. When the samples are cooled below T_C, the spontaneous magnetization splits the $3d$ up- and down-spin bands of the TM, thereby increasing the antibonding character which results in lattice expansion. Contributions from spin fluctuations of local moments on the TM ions could also be responsible for the deviation from Vegard's law. The effect of these are felt at temperatures below as well as above T_C. For example, in the case of Fe, it has been established that the local moments on Fe ions persist even well above T_C.[11]

Combining these two effects, it could be inferred that, in compounds with $x=0$, 0.5, and 1.0, for which the T_C values are well above RT, the deviation from Vegard behavior is a combined effect of band polarization and spin fluctuations. In the case of $x=1.5$, for which the T_C value is 270 K, probably the deviation could be mainly due to spin fluctuations of the local moments. In the case of $x=2.0$, Ni does not possess any magnetic moment and therefore, there are no contributions to MVE.

In addition to MVE, another factor which might bring about deviation of lattice parameters from Vegard behavior is charge transfer. In $Zr(Co_{1-x}Ni_x)_2$ system, which remains paramagnetic even up to very low temperatures, it has been reported that the deviation of lattice parameters is very small and is due to charge transfer.[14] Therefore, in the $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$ system, it could be inferred that the deviation of lattice parameters are mainly due to MVE.

Figure 2 shows the variation in magnetization as a function of applied magnetic field up to a value of 10 kOe at RT for all compounds prepared. It could be seen that the magnetization at 10 kOe decreases with increase in Ni concentration. Very little hysteresis was observed in these compounds. Figures 3(a)–3(e) show the virgin magnetization as a function of applied magnetic field at 4.2 K. The maximum field available was only 10 kOe and this was not sufficient to saturate the samples. In the case of $x=0$ [Fig. 3(a)], with increase in magnetic field, the magnetization changes very little up to about 1.5 kOe, above which it follows the usual ferromagnetic behavior. When the field was lowered from 10 kOe, a hysteresis of about 60 emu/g was observed. A similar behavior was also observed in samples with $x=0.5$, 1.0, and 1.5 as could be seen from Figs. 3(b)–3(d), respectively. For $x=2.0$, very little hysteresis was observed.

These results at 4.2 K indicate that there is a minimum field required to move the domain walls in the material. In high anisotropy systems, thickness of domain walls could be as small as a few lattice parameters.[15] Due to the presence of narrow domain walls, there is a difference between the domain wall energy when the position of the wall is between

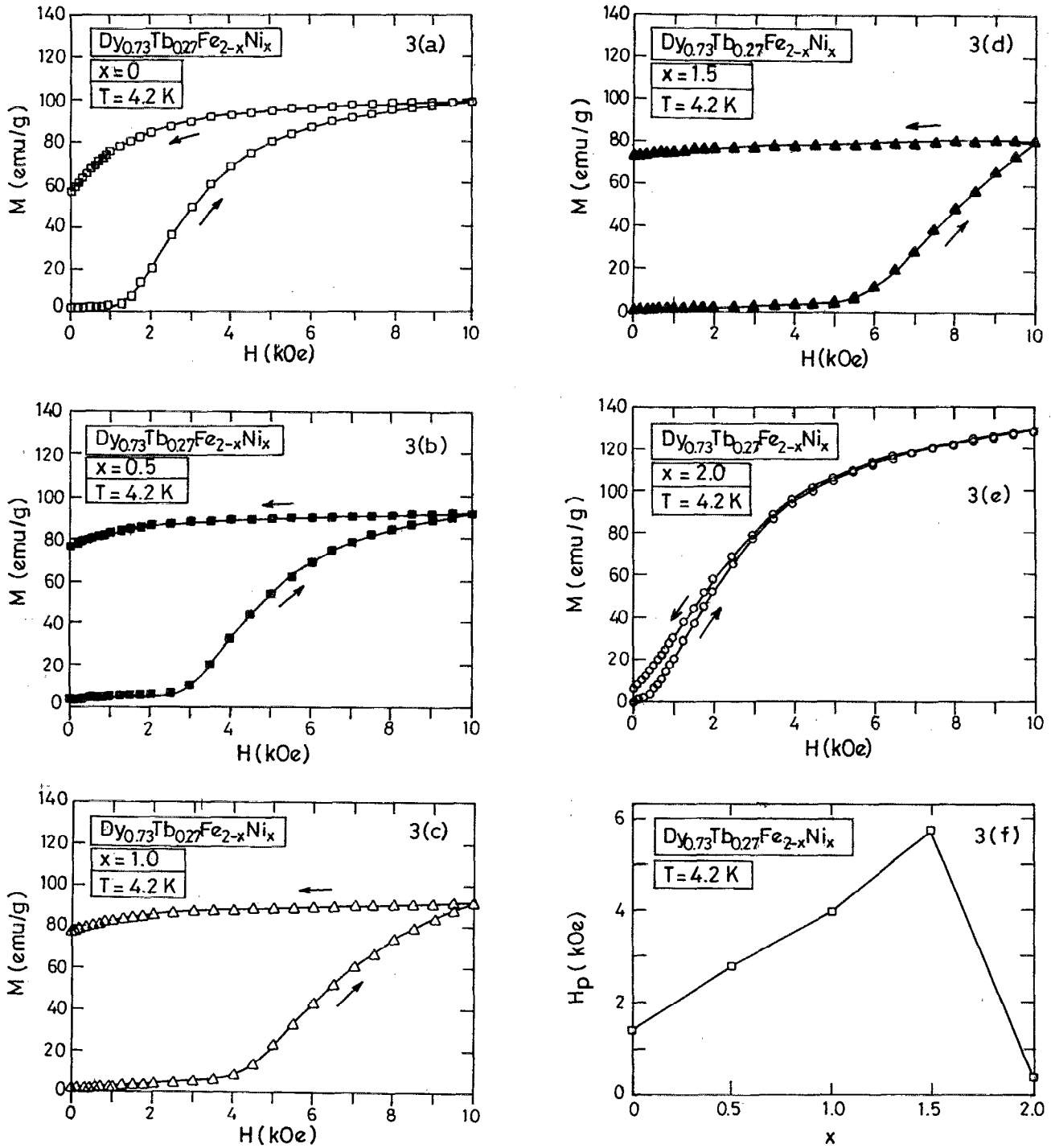

FIG. 3. (a)–(e) Variation in magnetization with applied magnetic field for various values of x at 4.2 K of $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$. (f) Variation in propagation field (H_p) with x at 4.2 K.

two atomic planes and that when the domain wall is at the atomic plane. This results in formation of energy barriers to the motion of the domain walls. Thus, the wall energy along the z direction in the crystal is given by the relation

$$V(z)=V(0)+\Delta V \sin(2\pi z/D), \tag{1}$$

where $V(0)$ is the wall energy at the minima of the potential energy curve, ΔV, the energy barrier and D, the interplanar distance.

A minimum field, known as propagation field (H_p), is required to reduce the height of these energy barriers to zero and therefore, the magnetization varies very little till the propagation field is reached. Above this field, the domain walls can easily propagate through the material and the usual ferromagnetic behavior of magnetization is achieved. When

the applied field is reduced, the energy barriers appear again. As a result, the walls get pinned to their present positions giving large hysteresis.

Figure 3(f) shows the H_p values as a function of x. These values increase linearly with Ni concentration up to $x=1.5$ and then fall to a small value for $x=2.0$. It has been established that the H_p is an increasing function of the ratio between anisotropy and exchange coupling constants.[15] The present results indicate that this ratio increases with x up to $x=1.5$ and decreases to a small value for $x=2.0$. Since the anisotropy in these systems is mainly due to the RE ions,[16] it can be assumed that the anisotropy at 4.2 K remains constant up to $x=1.5$. The decrease in T_C with x as shown in Fig. 5 indicates that the strength of exchange interaction decreases with x. Therefore, the H_p value increases up to $x=1.5$. In the case of $x=2.0$, the significantly low value of H_p shows that the anisotropy is reduced considerably.

These kind of pinning phenomena have been reported in $REFe_{2-x}Al_x$ and $REFe_{2-x}Ga_x$ systems.[17–19] A H_p value of 12 kOe has been observed in $DyFe_{1.4}Al_{0.6}$ while this value is 14 kOe in $DyFe_{1.6}Ga_{0.4}$. In the present investigation, the H_p value of 2.8 kOe obtained for $Dy_{0.73}Tb_{0.27}Fe_{1.5}Ni_{0.5}$ is much smaller than the values obtained for Al and Ga substituted samples. These results indicate that the pinning effect is more pronounced on Al and Ga substitution than on Ni substitution. When Ni replaces Fe, the rigid band approximation for the $3d$ band is valid. Therefore, the electronic structure remains the same for all the samples. On the other hand, when Fe is replaced by Al or Ga, the rigid band approximation is no longer valid and hence, the electronic structure is affected considerably. This will result in larger variations in exchange and anisotropy. Thus, a pronounced pinning effect is observed in Al or Ga substituted samples whereas the pinning effect is much less in the present system.

The temperature dependence of magnetization at an applied field of 10 kOe is shown in Fig. 4. The inset shows the behavior of the same curves in the vicinity of maxima on an expanded scale. The occurrence of these maxima are also due to the same domain wall pinning phenomena described earlier. It has been established that the anisotropy in $REFe_2$ alloys decreases as the temperature is increased.[1] When the temperature is increased from 4.2 K, H_p starts decreasing due to decrease in anisotropy and thermal excitation. As a consequence, the magnetization at 10 kOe starts increasing towards the saturation value of the specimen even though its temperature is increased. Thus, an increase in magnetization with temperature is observed. Above a certain temperature, the domain wall pinning effects become less pronounced and hence, the temperature dependence of magnetization begins to follow the usual ferromagnetic behavior.

T_C values determined from magnetization studies are shown in Fig. 5. These values decrease monotonically with increasing Ni content. Similar variations in T_C values have also been observed in $RE(Fe_{1-x}Ni_x)_2$ and $Dy_{0.73}Tb_{0.27}(Fe_{1-x}Ni_x)_{1.9}$ systems.[5,20,21] On the contrary, the T_C values increase up to $x=0.25$ and then decrease less rapidly in the case of $RE(Fe_{1-x}Co_x)_2$ systems.[6,7,22]

The results of electrical resistivity studies carried out in the temperature range 16–750 K are shown in Fig. 6. The T_C

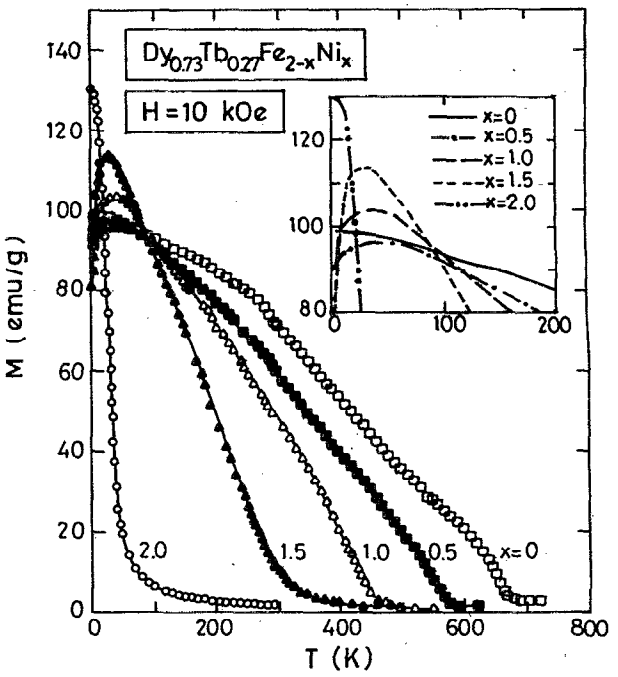

FIG. 4. Temperature dependence of magnetization at an applied field of 10 kOe for various x of $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$. The inset shows the behavior in the vicinity of maxima.

values obtained from these resistivity curves are plotted along with the T_C values obtained from magnetization measurements as shown in Fig. 5. A good agreement between these two measurements is observed. The inset in Fig. 6 shows the anomaly observed for $x=2.0$ in the vicinity of its T_C. This observation is similar to the one reported by us earlier in the $Ho_{0.85}Tb_{0.15}Fe_{2-x}Ni_x$ system.[5] When the sample is cooled from temperatures above T_C, critical fluctuations due to short-range spin ordering of RE's start increasing. Thus, the electrical resistivity due to the effect of

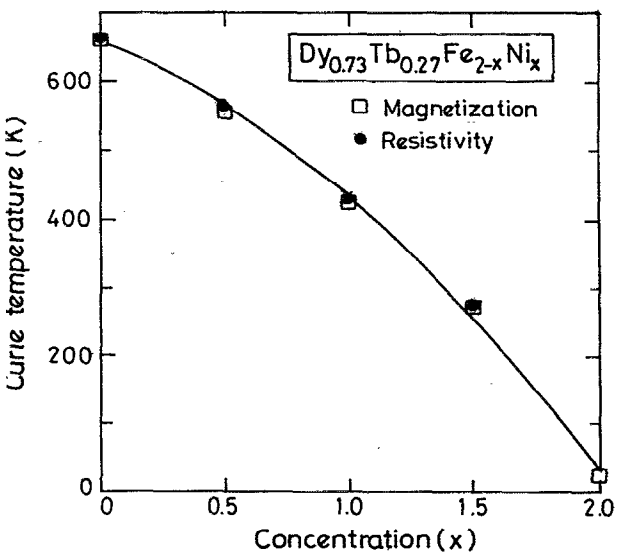

FIG. 5. Curie temperature as a function of x obtained from magnetization and electrical resistivity measurements on $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$.

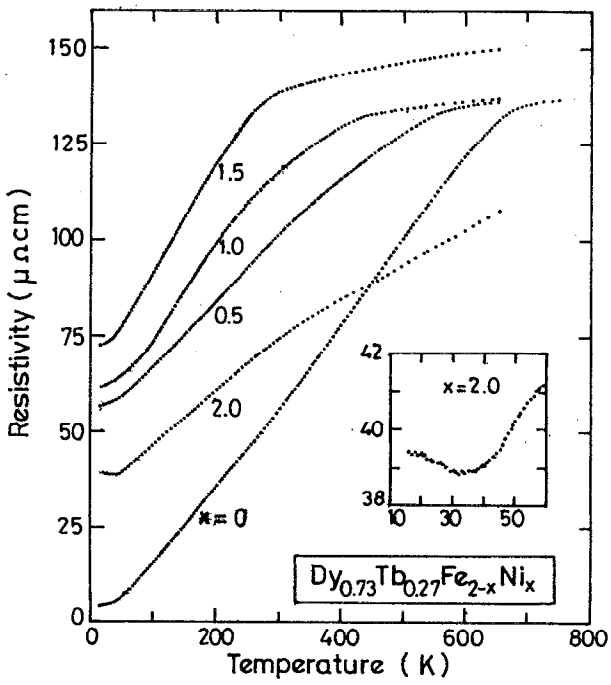

FIG. 6. Temperature dependence of electrical resistivity for various values of x of $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$. The inset shows the anomaly observed for $x=2$ in the vicinity of T_C.

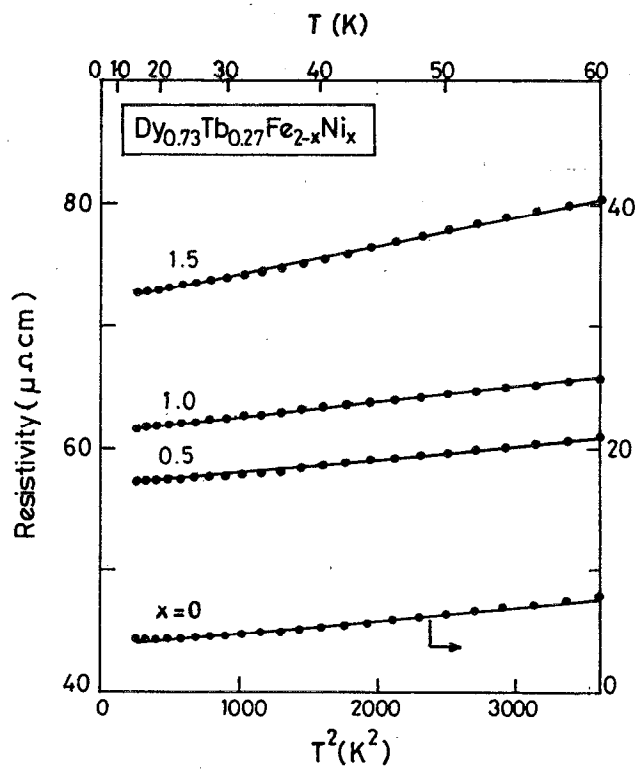

FIG. 7. Temperature dependence of electrical resistivity at low temperatures. The solid lines are the least-squares fit according to Eq. (3).

s-f exchange interaction between the conduction electrons and the localized f electrons near the magnetic transition starts increasing with decrease in temperature and therefore, the minimum is observed.[23,24] For $x<2$, the Fe-Fe interactions are stronger and therefore, the magnitude of resistivity arising due to these fluctuations in the s-f exchange interaction is negligible.

At low temperatures, the total resistivity is given by[25,26]

$$\rho(T)=\rho_0+aT^2\exp(-\Delta/kT)+bT^5, \tag{2}$$

where ρ_0 is the residual resistivity, k is the Boltzmann constant while a and b are constants. The second term in this expression is due to the electron-spin wave scattering. For systems possessing large magnetic anisotropy, a minimum energy Δ is required to excite a spin wave in the presence of an anisotropy field. The third term is due to the electron-phonon scattering. When the fit was carried out using Eq. (2) for the experimental data at low temperatures, the values of b and Δ obtained were found to be very small. Therefore, neglecting the contributions from these terms, the above expression could be rewritten as

$$\rho(T)=\rho_0+aT^2. \tag{3}$$

The experimental data fitted using Eq. (3) are shown in Fig. 7.

The scattering of conduction electrons by spin waves depends upon the energy spectrum of the spin waves. The spin wave spectrum for the ferrimagnets comprises of one acoustic and many optical modes of excitation and each mode will experience a different amount of anisotropy. The acoustic mode is the lowest energy mode which could be excited at low temperatures and the population of the optical modes would be negligible. From the spin wave spectrum obtained for $HoFe_2$ it could be noted that the acoustic branch arising due to RE-Fe exchange interaction has a Δ value less than 1 meV at 10 K, which is very small.[27] The optical modes observed possess large values of Δ. The small values of Δ obtained in our results indicate that only the acoustic mode due to RE-Fe exchange contributes to resistivity at low temperatures.

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