

^{57}Fe Mössbauer investigations on the $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems

M. Senthil Kumar, K. V. Reddy, and K. V. S. Rama Rao*

Magnetism and Magnetic Materials Laboratory, Department of Physics, Indian Institute of Technology, Madras 600 036, India

T. P. Das

Department of Physics, State University of New York at Albany, Albany, New York 12222

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^{57}Fe Mössbauer studies were carried out on the $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems ($x=y=0, 0.5, 1.0, 1.5$). The spectra were recorded in the temperature range 18–675 K. The variation of hyperfine field at 18 K with Ni concentration showed a maximum at $x=0.5$ as well as $y=0.5$ which has been attributed to the narrowing of the $3d$ bands by the substitution of Ni. The Curie temperatures were estimated for all values of x and y from the temperature dependence of the average hyperfine field, which followed the Brillouin function. A monotonic decrease in Curie temperature with Ni concentration is observed and this is attributed to the decrease in net transition-metal sublattice moment. From the Mössbauer spectra, spin reorientation transitions have been observed for compositions with $x=y=0, 0.5$, and 1.0 .

I. INTRODUCTION

Studies on cubic Laves phase $R\text{Fe}_2$ (R is a rare earth) alloys, particularly the anisotropy-compensated $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_2$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_2$, are of great importance owing to their potential application as magnetostriuctive transducer elements.^{1,2} The binary $R\text{Fe}_2$ systems are found to have large magnetostriction associated with large magnetic anisotropy and therefore high magnetic fields are required to saturate the materials. The sign of the first-order magnetocrystalline anisotropy constant K_1 is negative for TbFe_2 and is positive for DyFe_2 and HoFe_2 . Therefore it is preferable to alloy materials with the same sign of magnetostriction but with opposite signs of anisotropy. Clark and Belson³ and Koon *et al.*⁴ have found that alloys with the compositions $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_2$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_2$ possess low magnetic anisotropy (with very low K_1) while retaining large magnetostriction which can be used for practical applications.^{1,2} These alloys belong to the category of $R\mathcal{M}_2$ (\mathcal{M} is a transition metal) systems possessing the $C15$ -type cubic Laves-phase structure. The striking feature of the $R\mathcal{M}_2$ systems is the presence of localized $4f$ electrons and itinerant $3d$ electrons. The density of states (DOS) of the $3d$ band plays a crucial role in determining the magnetic state of the $3d$ electrons. The Fermi level is located in a delicate position where it can display diverse magnetic properties. In these systems, Fe carries a magnetic moment, Co has an induced moment due to the large molecular field of R , whereas Ni has no moment due to a completely filled $3d$ band. Thus $R\text{Fe}_2$ and $R\text{Co}_2$ are ferrimagnetic while $R\text{Ni}_2$ is ferromagnetic.^{5,6} ^{57}Fe Mössbauer studies provide microscopic information about the electronic structure of these materials and a change in magnetic properties can reveal the details of the DOS at the Fermi level.

The structural properties of various $R\text{Fe}_{2-x}\text{Ni}_x$ systems have been investigated by Mansey, Raynor, and Harris⁷ and Christopher, Piercy, and Taylor.⁸ An earlier report on $\text{DyFe}_{2-x}\text{Ni}_x$ using ^{57}Fe Mössbauer studies and

the temperature dependence of the reciprocal susceptibility in the paramagnetic region by Burzo⁹ has revealed that the hyperfine field at 4.2 K and the effective Fe moment determined from the temperature dependence of the reciprocal susceptibility decrease with increasing Ni concentration. On the other hand, Muraoka, Shiga, and Nakamura¹⁰ have found that in $\text{YFe}_{2-x}\text{Ni}_x$ there is an increase in the Fe hyperfine field at 4.2 K with increasing Ni concentration up to $x=0.2$. Their results, obtained from magnetization studies, have also indicated an increase in Fe moment even though the net \mathcal{M} sublattice moment decreases.

Recently we have investigated the effect of Ni on the structural, magnetic, and electrical properties of $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems.^{11–13} It was found that in both systems there is a decrease of lattice parameter with increasing Ni concentration. This, change which did not follow Vegard's law, has been attributed to the presence of the magnetovolume effect. The temperature variation of magnetization carried out in the temperature range 4.2–750 K did not reveal the presence of any spin reorientation transition. The data at 4.2 K indicated the domain-wall pinning effect which is very prominent in the case of $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ compared to $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$. Electrical resistivity studies carried out in the temperature range 18–750 K showed anomalies corresponding to the Curie temperatures (T_C). In continuation of these studies we present here detailed investigations of ^{57}Fe Mössbauer studies carried out on $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems in the temperature range 18–675 K.

II. EXPERIMENTAL DETAILS

The samples of $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ with $x=y=0, 0.5, 1.0$, and 1.5 were prepared by arc-melting the individual elements followed by annealing. The details of preparation of the

samples were reported previously.^{11,12} Powder x-ray diffractograms were taken to confirm single-phase formation. The spectra were recorded using a CMTE Mössbauer spectrometer with a $^{57}\text{Co}:\text{Rh}$ source kept at room temperature (RT). Low-temperature experiments were performed in the temperature range 18–300 K using a closed-cycle helium refrigerator.¹⁴ Experiments were conducted by keeping the samples in a helium-exchange gas chamber which was fixed above the cold-head second stage. The samples were kept at the bottom of a sample holder fixed to a rigid support. This rigid support is isolated from the cold-head support by means of a bellow assembly in order to avoid the transmission of vibrations produced at the cold head to the sample holder. Mylar foils were used as windows in both the sample chamber and the outer vacuum shroud. A carbon-glass resistor was used as the temperature sensor and the temperatures were maintained within ± 0.1 K. High-temperature experiments were carried out using an oven assembly with beryllium windows. The sample chamber was evacuated continuously using a diffusion pump with an ultimate vacuum of 10^{-6} Torr. The samples were mixed with boron nitride and sandwiched between two beryllium disks. A Chromel-Alumel thermocouple was used as temperature sensor and the temperatures were maintained within ± 1 K.

III. RESULTS, ANALYSIS OF SPECTRA, AND DISCUSSION

The Mössbauer spectra of the $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems for $x=y=0, 0.5, 1.0$, and 1.5 at different temperatures are shown in Figs. 1 and 2. The spectra show different features depending on the direction of easy magnetization. The quadrupolar split doublets in the paramagnetic state are also shown.

In the RFe_2 compounds possessing the $C15$ -type cubic Laves-phase structure, the Fe ions are situated in a corner-sharing network of regular tetrahedra and all the Fe ions are crystallographically equivalent. The site symmetry of Fe ions is $\bar{3}m$ and the threefold axes lie along the $[111]$ directions. The electric-field gradient (EFG) tensor at the Fe nuclei is axially symmetric and the axes of these gradients are parallel to the local threefold axes of symmetry. Depending on the angle θ between the directions of easy magnetization and the electric-field gradients, three different types of spectra can be obtained.¹⁵ The quadrupole splitting for each site differs from the others due to the presence of $\cos^2\theta$ term in the quadrupole Hamiltonian. With the direction of easy magnetization along the $[100]$ axis, the angle θ formed between the directions of easy magnetization and the EFG is $54^\circ 44'$ for all the Fe ions and a simple six-line

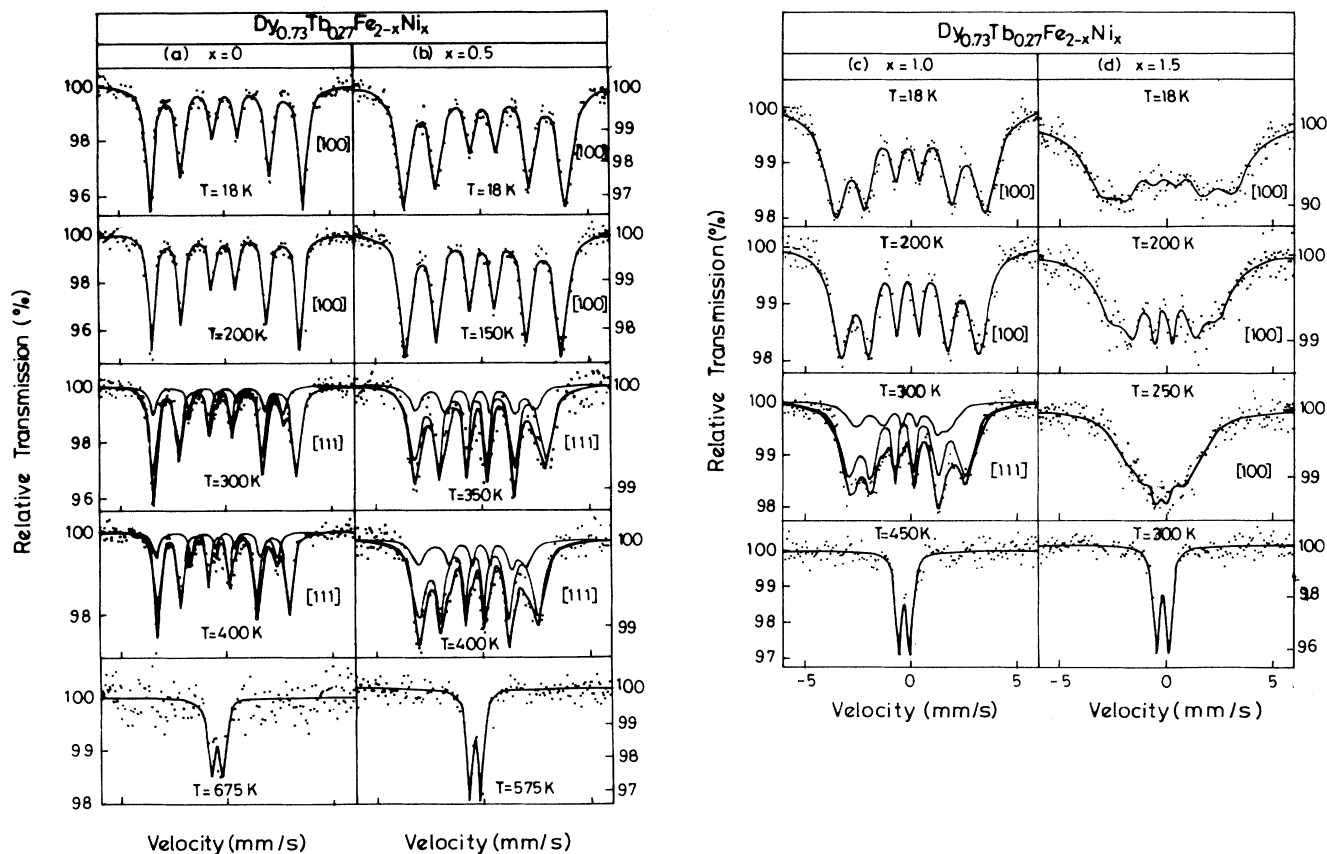


FIG. 1. Mössbauer spectra of the $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ system for (a) $x=0$, (b) $x=0.5$, (c) $x=1.0$, and (d) $x=1.5$ at different temperatures.

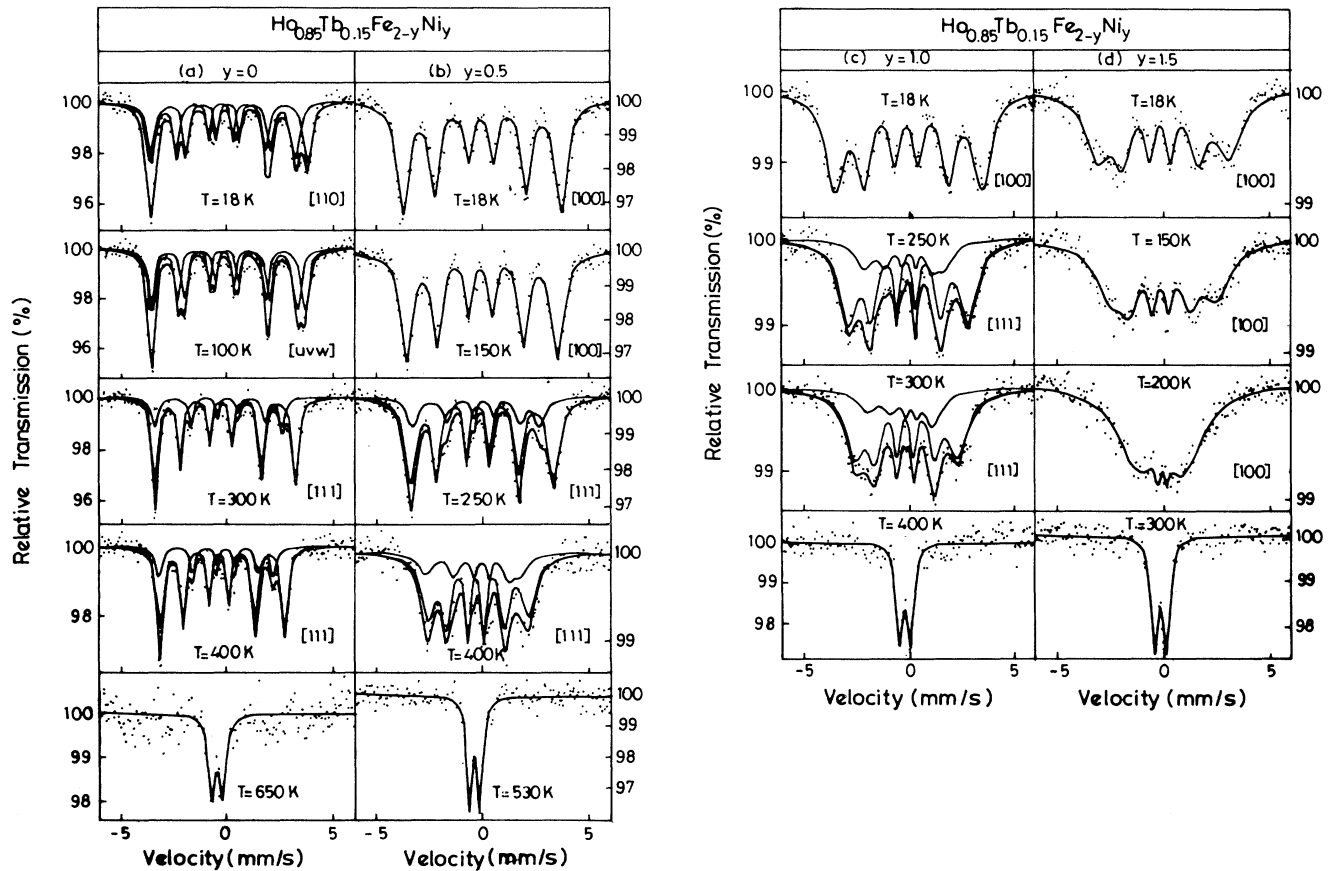


FIG. 2. Mössbauer spectra of the $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ system for (a) $y=0$, (b) $y=0.5$, (c) $y=1.0$, and (d) $y=1.5$ at different temperatures.

spectrum is expected. When the easy magnetization axis is along the [111] direction, two magnetically inequivalent Fe sites, due to different θ values of $70^\circ 32'$ and 0° , with relative population ratio 3:1 exist, giving rise to a spectrum which is a superposition of two six-line patterns with the intensity ratio 3:1. In the case of the easy magnetization direction along [110], two magnetically inequivalent iron sites with a population ratio 1:1 due to θ values of $35^\circ 16'$ and 90° are present. Thus, depending on the nature of the resolved spectra, the direction of easy magnetization is deduced. When the spectra did not exhibit any one of the above-mentioned features, the direction of easy magnetization was designated as [uvw]. In order to extract various parameters, the spectra corresponding to [uvw] were fitted to [111], except for the transition region 75–225 K in $y=0$ where they were fitted to [110].

The variations of the average hyperfine field (H_{hf}) at 18 K with x and y are shown in Figs. 3(a) and 3(b). The H_{hf} values at 18 K are 224 and 234 kOe for $x=0$ and 0.5, respectively. Above $x=0.5$, this value decreases rapidly. Similarly, the values of H_{hf} at 18 K are 222 and 232 kOe for $y=0$ and 0.5, respectively, and a rapid decrease is observed above $y=0.5$. It was established that there is a direct proportionality between H_{hf} and the magnetic moment of Fe and the proportionality factor is 150

kOe/μ_B .¹⁶ The H_{hf} values are converted to magnetic moments using this proportionality factor. These values are also shown in Fig. 3.

It can be seen from Fig. 3 that the magnetic moment at the Fe site in both systems reaches a maximum value at $x=0.5$ and $y=0.5$. Burzo⁹ has reported that there is a decrease in H_{hf} at 4.2 K with x in $\text{DyFe}_{2-x}\text{Ni}_x$, and the decrease is less rapid in the Fe-rich region whereas these values fall rapidly in the Ni-rich region. The results obtained from the temperature dependence of the inverse susceptibility have also indicated the same behavior. In contrast, the studies by Muraoka, Shiga, and Nakamura¹⁰ on $\text{YFe}_{2-x}\text{Ni}_x$ up to $x=0.2$ showed an increase in H_{hf} at 4.2 K with x . Magnetization studies also showed that there is an increase in Fe moment with x even though the net \mathcal{M} sublattice moment decreases. They attributed this observation to incomplete polarization of the 3d bands due to weak ferromagnetism in terms of the Stoner model. The results of Ref. 10 and the results of the present study clearly indicate that there is an increase of Fe moment with the substitution of Ni. The reason for this increase in moment can be understood by comparing these results with those obtained on the $\text{YFe}_{2-x}\text{Co}_x$ system.

A maximum in the magnetization at 4.2 K as a function of x in the $\text{YFe}_{2-x}\text{Co}_x$ system was reported earlier by Piercy and Taylor¹⁷ and was explained based on the

rigid-band model. They suggested that the Fermi level in YFe_2 lies at the minimum of the DOS of the down-spin subband and at the maximum of the DOS of the up-spin subband. As electrons are added to the band on the substitution of Co, the increase in the number of up-spins is more than that of the down-spins. Thus the net $3d$ moment starts increasing with increasing Co concentration. Above a certain Co concentration the DOS at the Fermi level in the down-spin band will be higher than that of the up-spin band and hence a further addition of electrons would result in a reduction of the moment. In such a case, the electronic specific heat coefficient γ should fall appreciably with increasing Co concentration up to the maximum observed and then should increase with further increase in Co concentration. But the γ values observed by Muraoka, Shiga, and Nakamura¹⁸ did not change appreciably with x in $YFe_{2-x}Co_x$ in the region where the maximum in magnetic moment was observed. This suggests that the structure of the DOS of the $YFe_{2-x}Co_x$

system is different from what has been proposed by Piercy and Taylor.¹⁷

Eriksson *et al.*,¹⁹ from their band-structure calculations, have attributed the observation of a maximum in $YFe_{2-x}Co_x$ to the narrowing of the $3d$ bands due to the increase in atomic number. As the atomic number increases from 26 in the case of Fe to 27 in the case of Co, an increase in the localization of electrons occurs. Thus the $3d$ bandwidth decreases with gradual substitution of Co in place of Fe. On substitution of Co, a gradual filling up of the $3d$ band takes place. The difference between the number of up- and down-spins increases until the spin-up subband is completely filled, resulting in an increase of moment. After this, the difference between the number of up- and down-spins decreases with further filling up of the down-spin band, resulting in the decrease of the net $3d$ moment. Thus a maximum is observed in the moment versus composition curve.

In the present investigation, Fe is replaced by Ni, whose atomic number is 28. As discussed in the case of the $YFe_{2-x}Co_x$ system in the present systems also the $3d$ bandwidth decreases with increasing Ni concentration due to the localization of electrons. The maximum occurs in both systems around $x=y=0.5$, in the present cases. Therefore we attribute the observation of maxima in the variation of H_{hf} at 18 K with Ni concentration in the present investigation to the decrease in $3d$ bandwidth.

Due to the localization of electrons, there will be a decrease in the ionic radii which will lead to a decrease in lattice parameter with increasing Ni concentration. Our earlier studies on these systems showed a decrease in the lattice parameter with increasing Ni concentration in both systems.^{11,12}

The temperature dependence of the average H_{hf} for various x and y is shown in Figs. 4(a) and 4(b). The solid lines represent the fit using the Brillouin function $B_J(x)$ where $x = AH_{hf}(T)/T$. Here A is an empirical constant. The J value of the Fe ions is fixed as 0.75, which is found to be appropriate in RFe_2 systems.²⁰ The T_C values were determined by imposing the condition $H \rightarrow 0$ when $T \rightarrow T_C$, in the Brillouin function.^{21,22} The T_C values estimated from these curve fittings agree reasonably well (within 3–4%) with the values obtained from magnetization and resistivity measurements.^{11,12} These T_C values are plotted as a function of Ni concentration as shown in Figs. 5(a) and 5(b). A monotonic decrease in T_C is observed in both systems. On the other hand, an initial increase up to approximately $x=0.5$ and a further decrease above this x value is found in various $RFe_{2-x}Co_x$ systems.^{17,23–25}

According to the spin fluctuation theory of Mohn and Wohlfarth,^{26,27} the T_C values in $R\mathcal{M}_2$ systems can be written as

$$T_C \propto \frac{M_0^2}{\chi_0} \quad (1)$$

where

$$\chi_0^{-1} = \left[\frac{1}{2N_{\uparrow}(E_F)} + \frac{1}{2N_{\downarrow}(E_F)} - I \right] \frac{1}{2\mu_B^2}$$

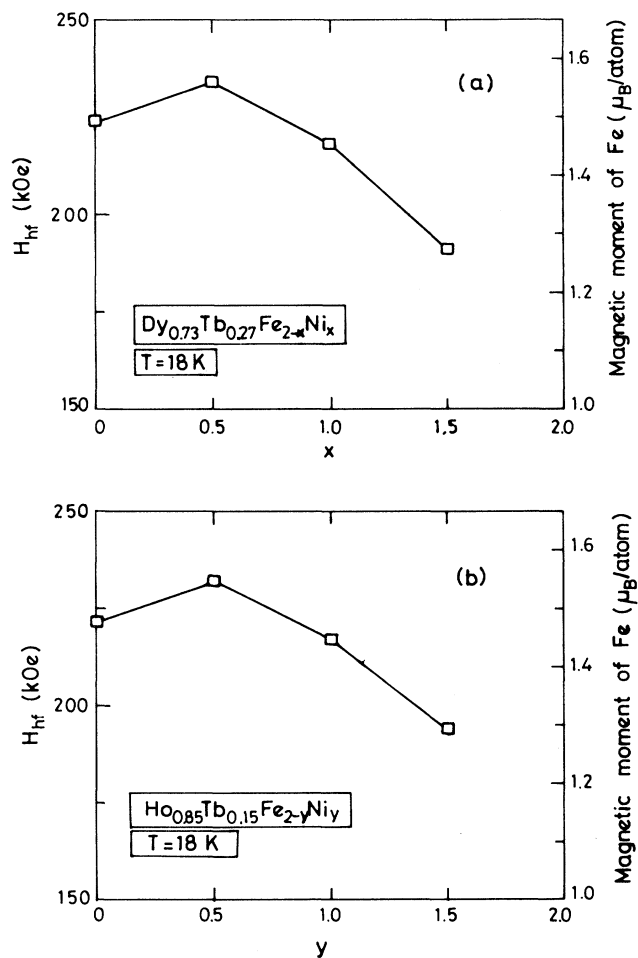


FIG. 3. Variation of hyperfine field at 18 K with Ni concentration in (a) $Dy_{0.73}Tb_{0.27}Fe_{2-x}Ni_x$ and (b) $Ho_{0.85}Tb_{0.15}Fe_{2-y}Ni_y$. The Fe magnetic-moment variation is calculated based on the linear relationship between H_{hf} and moment (Ref. 16).

Here χ_0 is the enhanced susceptibility, M_0 is the magnetic moment per atom at 0 K, $N_\uparrow(E_F)$ and $N_\downarrow(E_F)$ are the density of states at the Fermi level, and I is the Stoner parameter. This relation was derived based on the fact that the M - M interaction forms the major factor which determines the values of T_C in these systems. Since the rigid-band model can be applied to systems containing Ni and Co, one can assume that the change in χ_0 is nearly the same in both Ni- and Co-substituted systems except that the d -band filling rate is higher in Ni systems than in Co systems. But the variation in magnetic moment of Fe with composition shows a maximum with Ni as well as Co substitutions. In the case of Co-substituted systems, the total magnetic moment of the M sublattice is the sum $(2-x)M_0^{\text{Fe}} + xM_0^{\text{Co}}$ whereas it is only $(2-x)M_0^{\text{Fe}}$ for Ni-substituted systems, since Ni is nonmagnetic. The results of Piercy and Taylor¹⁷ on $\text{YFe}_{2-x}\text{Co}_x$ showed a maximum in the variation of net M sublattice moment with x . In the $\text{YFe}_{2-x}\text{Ni}_x$ system, the net M sublattice moment decreases with increasing x as reported by Muraoka, Shiga, and Nakamura.¹⁰ Thus, incorporating the net M sublattice moments in relation (1), the maxima in T_C

observed in $\text{RFe}_{2-x}\text{Co}_x$ systems and a monotonic decrease of T_C with x in $\text{RFe}_{2-x}\text{Ni}_x$ systems can be understood.

From the spectra obtained at various temperatures for all the samples, spin orientation diagrams are constructed for each system and are as shown in Figs. 6(a) and 6(b). In the case of the $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ system, the spins are oriented along [100] at 18 K for all the Ni compositions. For $x=0$, a transition from [100] to [111] takes place in the temperature interval 230–280 K. In the case of $x=0.5$ and 1.0, similar spin reorientation from [100] to [111] takes place around the same temperature range. However, in these cases, the widths of the transitions are large compared to that for $x=0$. No spin reorientation is observed for $x=1.5$. In the case of the $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ system the spins are oriented along [100] at 18 K for $y=0.5, 1.0,$ and 1.5 . In the case of $y=0$, the spin orientation direction is [110] at 18 K, a reorientation takes place as the temperature increases, and above 250 K the orientation is [111]. For $y=0.5$ and 1.0, a spin reorientation transition from [100] to [111] is observed. No reorientations are observed for $y=1.5$. In the case of the $\text{Dy}_{0.7}\text{Tb}_{0.3}\text{Fe}_{2-x}\text{Co}_x$ system, as reported by Segnan and Deriu,²⁸ the spin orientation at 4.2 K is

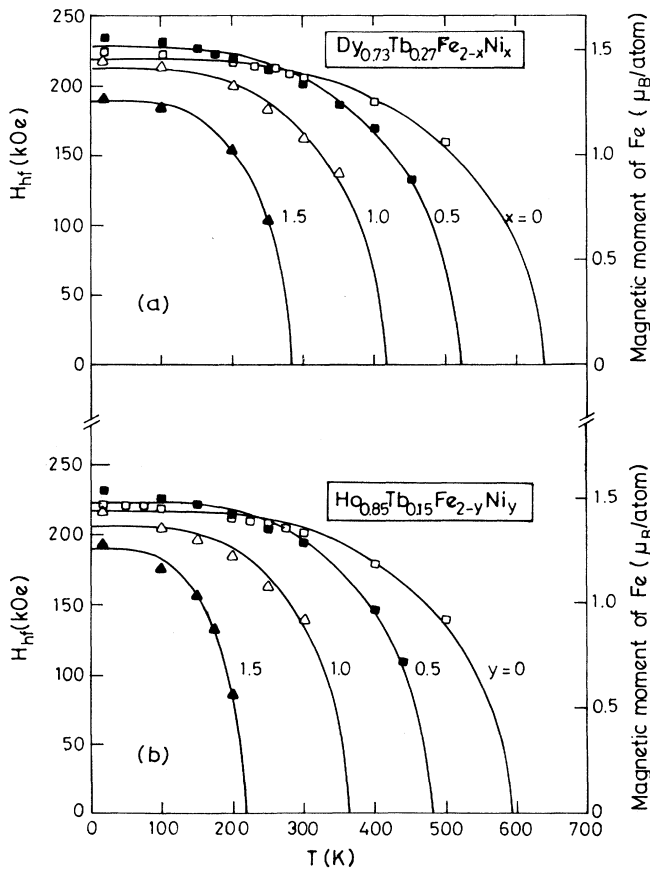


FIG. 4. Temperature dependence of average hyperfine field for various Ni concentrations in (a) $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and (b) $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$. The solid lines represent the curve fitting using the Brillouin function. The Fe moment variation is calculated based on the linear relationship between H_{hf} and moment (Ref. 16).

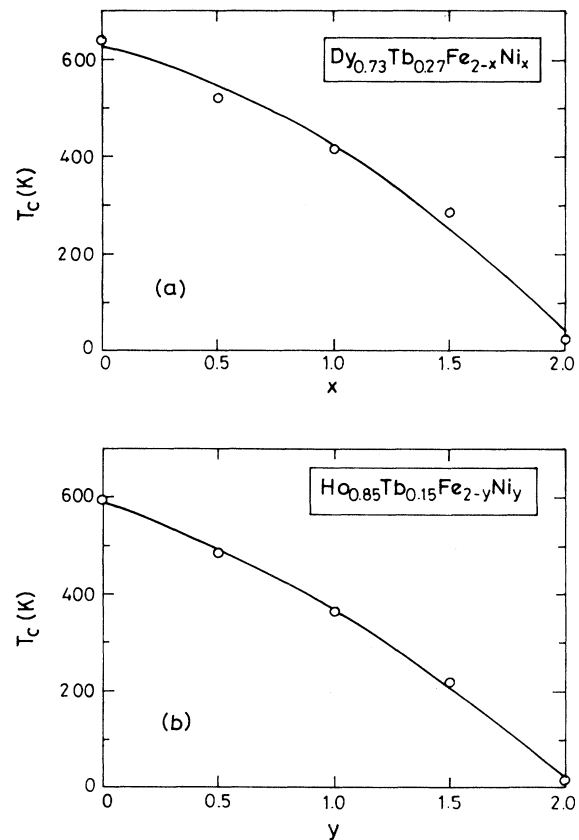


FIG. 5. Variation of T_C as a function of Ni concentration in (a) $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and (b) $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems. The T_C values for $x=2.0$ and $y=2.0$ were taken from Ref. 12 and Ref. 11, respectively.

[100] for $x=0-0.6$, and for $x=0.9$ and above the spins are oriented along [111].

These orientations of spins along a particular direction at a given temperature depend on the crystalline electric fields acting on the $4f$ electrons of the R ions. Due to the crystalline fields acting on the $4f$ electrons, the degeneracy of the J multiplet is removed, resulting in the splitting of levels, and when the exchange field splits these levels further the anisotropy is realized. Thus the anisotropy is determined by the values of crystal-field parameters and the exchange splitting. At any temperature, the anisotropy is determined by the population of these levels and the spins will orient themselves along the direction so that the free energy of the system is minimum. Even though the anisotropy is determined mainly from the R ions, the M ions also influence the crystal-field interactions by a small additional contribution to the anisotropy and indirectly influence these crystal-field interactions through the changes in lattice parameter. Therefore the changes in the spin orientations in the present systems are due to changes in the crystal-field interactions and exchange splitting introduced by the Ni ions.

Mössbauer spectra indicate the occurrence of spin re-orientation transitions. However, these occur over a wide temperature range and therefore did not reflect in our magnetization studies reported earlier.^{11,12}

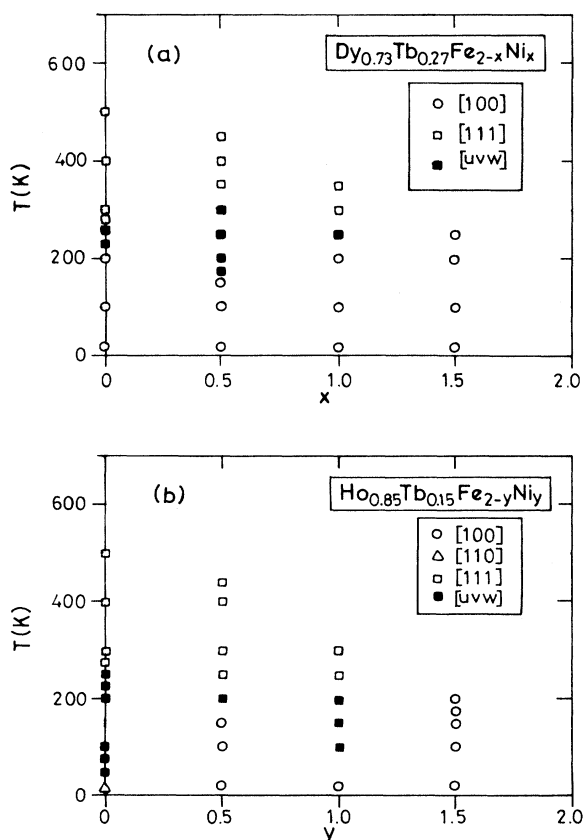


FIG. 6. Spin orientation diagrams of (a) $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and (b) $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems.

Figure 7 shows the variation of isomer shift (with respect to natural iron at RT) with temperature for various x and y in both systems. The solid lines represent the calculated variation of isomer shift with temperature according to the Debye model of second-order Doppler shift which is given as²⁹

$$\delta(T) = \frac{-9k\Theta_D}{2Mc} \left[\frac{1}{8} + \left(\frac{T}{\Theta_D} \right)^4 \int_0^{\Theta_D/T} \frac{x^3 dx}{(e^x - 1)} \right], \quad (2)$$

where k is the Boltzmann constant, M the mass of the ^{57}Fe nucleus, c the velocity of light in vacuum, and Θ_D the Debye temperature. This expression with a proper additive constant was used for fitting the data. Since the number of experimental data available on the samples with various x and y is smaller, the Θ_D values which can be deduced from the fit may not be accurate. Therefore we assumed that the Θ_D values do not change with x as well as y and fitted a single curve to all the data available in each system under investigation. This assumption is valid, since the Θ_D values obtained by Muraoka, Shiga, and Nakamura¹⁸ do not change with x up to 1.0 in the $\text{YFe}_{2-x}\text{Co}_x$ system and above $x=1.0$ an increase is observed which is due to the spin fluctuations of Co. In the

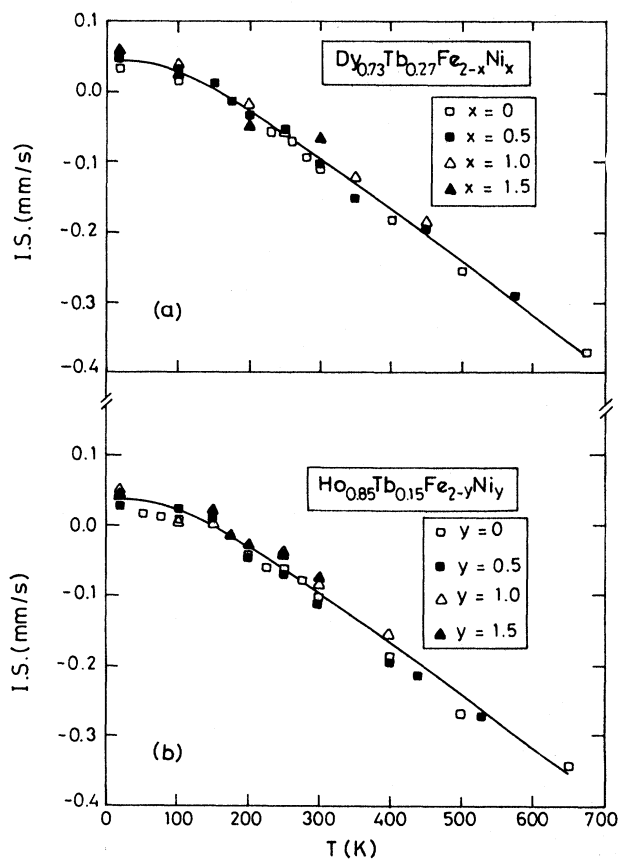


FIG. 7. Temperature dependence of isomer shift in (a) $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and (b) $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems for various x and y . The solid lines represent the fit based on the Debye model of second-order Doppler shift.

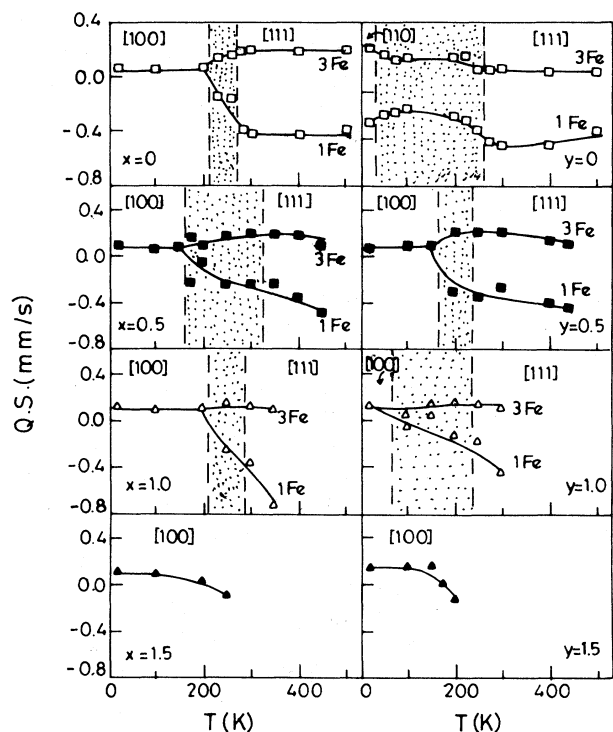


FIG. 8. Temperature variation of quadrupole splitting in $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems for various x and y . The dotted area indicates the transition region $[uvw]$.

present case, Ni is nonmagnetic and therefore such an increase in Θ_D is not expected. The values of Θ_D obtained from the fitting are 360 K for the $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ system and 390 K for the $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ system, which fall in the range of values reported for various RFe_2 compounds.^{30,31} The variation of isomer shift with the concentration of Ni is not significant in either system. On the other hand, the isomer shift values reported in $\text{YFe}_{2-x}\text{Al}_x$ showed a large change with x . This indicates a large change in the s -electron density, probably because of the different electronic configurations of Al and Fe ions.³²

Figure 8 shows the quadrupole splittings obtained for all the samples at various temperature. The figure clearly indicates the change in the quadrupole-splitting values when the samples undergo spin reorientations.

IV. CONCLUSIONS

Mössbauer studies were carried out on the $\text{Dy}_{0.73}\text{Tb}_{0.27}\text{Fe}_{2-x}\text{Ni}_x$ and $\text{Ho}_{0.85}\text{Tb}_{0.15}\text{Fe}_{2-y}\text{Ni}_y$ systems at various temperatures. A maximum in the variation of H_{hf} at 18 K with Ni concentration was found in both systems at $x=y=0.5$. These results indicate that the magnetic moment of Fe shows a maximum with change in Ni concentration in both systems and this is attributed to the narrowing of the $3d$ bands because of the localization of electrons with increasing atomic number. The increasing localization of electrons with increasing atomic number would also result in a reduction in the ionic radii. The decrease in lattice parameter with increasing Ni concentration observed by us earlier also indicates the localization of electrons.^{11,12} The temperature variation of H_{hf} for various samples followed the Brillouin function and the T_C values were estimated. These T_C values are in agreement with the values obtained from magnetization and electrical resistivity measurements reported by us earlier.^{11,12} The monotonic decrease in T_C with increasing Ni concentration in the present systems and a maximum in the variation of T_C with Co concentration observed in $\text{RFe}_{2-x}\text{Co}_x$ systems were attributed to the variation of net \mathcal{M} sublattice moments in the respective systems. From the spectra recorded at various temperatures, spin orientation diagrams were constructed for each system. In all the samples, at 18 K the spin orientation is along [100], except for $y=0$ for which the spin orientation is along [110]. With increase in temperature (with the exception of $x=y=1.5$) the spin orientation changes to [111] through $[uvw]$. These studies clearly indicate that the spin reorientation transitions in the present systems occur over a wide temperature range. Earlier studies on the magnetization in both systems did not show the presence of any spin reorientation transition.^{11,12} This could be due to the occurrence of the change in magnetization over a wide temperature range during the spin reorientation process and therefore no anomaly could be detected.

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*Author to whom all correspondence should be addressed.

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