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Role of Fe substitution on the anomalous magnetocaloric and magnetoresistance behaviour in $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ compounds

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Abstract

We report the magnetic, magnetocaloric and magnetoresistance results obtained from $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ compounds with $x = 0, 0.025$ and 0.05 . Fe substitution leads to an increase in the ordering temperature from 36 K for $x = 0$ to 124 K for $x = 0.05$. In contrast to the single sharp magnetocaloric effect (MCE) peak seen in TbNi_2 , the MCE peaks of the Fe-substituted compounds are quite broad. We attribute the anomalous MCE behaviour to the randomization of the Tb moments brought about by the Fe substitution. Magnetic and magnetoresistance results seem to corroborate this proposition. The present study also shows that the anomalous magnetocaloric and magnetoresistance behaviour seen in the present compounds is similar to that of $\text{Ho}(\text{Ni}, \text{Fe})_2$ compounds.

1. Introduction

The property of the magnetic materials to heat up or cool down when they are subjected to a varying magnetic field under adiabatic conditions is known as the magnetocaloric effect (MCE). Generally, the MCE is measured as an isothermal magnetic entropy change and/or an adiabatic temperature change. Materials with considerable MCE over a wide temperature span find applications as active materials in magnetic refrigerators. The discovery of a giant MCE in a few intermetallic compounds [1–3] has led to intense research towards the development of a magnetic refrigerator for room-temperature applications. However, apart from its potential in the near room-temperature region, magnetic refrigeration technology could play a vital role in cryogenic applications spread over a wide temperature range [4, 5]. For example, the principle of magnetic refrigeration is being used in devices like gas liquefiers, cryo-coolers etc [6]. Furthermore, some recent reports suggest that magneto-thermal properties associated

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with magnetic materials could also be exploited in the fabrication of heat pumps. Therefore, the search for novel magnetic materials and studies of their MCE behaviour are of great relevance today.

The manifestation of diverse magnetic properties by the rare earth (R)–transition metal (TM) intermetallic compounds in general and the occurrence of a giant MCE in materials such as $\text{Gd}_5(\text{Si}, \text{Ge})_4$ in particular have made R–TM compounds the natural probe for fundamental studies as well as for applications based on the MCE [1, 7–9]. Among the various R–TM intermetallics, RCo_2 (R = Er, Ho and Dy) compounds are known to exhibit a considerable MCE due to the first-order transition at their ordering temperature (T_C) [8]. Another series which is quite promising is RNiAl . The large table-like MCE observed in many compounds of this series is attributed to the presence of multiple magnetic transitions in this series of compounds [9]. Systems based on $\text{La}(\text{Fe}, \text{Si})_{13}$ also exhibit a significant MCE, enabling them to be considered as potential refrigerants [10]. Most of the materials studied so far show a considerable MCE close to their magnetic ordering temperatures, resulting in a single peak in the temperature variation of MCE plots. However, there exist a few materials, typical examples being GdMn_2 and Gd_3Al_2 , which show a double-peak MCE behaviour [11, 12]. In general, materials with close-by multiple transitions are of interest in the field of magnetic refrigeration since they may lead to a considerable MCE over a wide range of temperatures.

As part of the programme to develop novel magnetocaloric materials suitable for different temperature ranges and to study the correlation between the magnetism and the MCE, we have been focusing our studies on various RTM_2 Laves phase compounds [13–15]. Recently, we found that Fe substitution results in an anomalous MCE and magnetoresistance (MR) in $\text{Ho}(\text{Ni}, \text{Fe})_2$ system [15]. In the case of the MCE, the anomaly was manifested in the form of a broad double-peak structure in the Fe-substituted compounds. This behaviour was attributed to the randomization of the moments in the Ho sublattice, brought about by the Fe substitution, at low temperatures. The magnetization and MR results were found to corroborate the presumption of randomization. In order to ascertain whether this behaviour is linked to a particular rare earth, we have taken up another system, namely $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ (with $x = 0, 0.025$ and 0.05), and studied the MCE and MR, the results of which are presented in this paper.

2. Experimental details

Polycrystalline compounds of $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ with $x = 0, 0.025$ and 0.05 were prepared by arc melting the constituent elements of at least 99.9% purity in argon atmosphere. The ingots were melted several times to ensure homogeneity. The arc-melted ingots were then annealed at 800°C in argon-filled quartz ampoules for a week. The compounds were characterized using powder x-ray diffraction (XRD) patterns obtained at room temperature using $\text{Cu K}\alpha$ radiation. The lattice parameters were calculated using the Rietveld refinement of the XRD data. Magnetization measurements in the temperature range 2–240 K, and up to a maximum field of 50 kOe, were carried out using a vibrating sample magnetometer (VSM, Oxford Instruments). The ac magnetic susceptibility was measured using a physical property measurement system (PPMS, Quantum Design) in the temperature range 2–200 K, under zero bias field as well as in a bias field of 20 kOe. The amplitude of the ac field was 5 Oe and the frequency was 333 Hz. Heat capacity measurements, in the temperature range 2–280 K and in fields up to 50 kOe, were performed using the relaxation method in the PPMS. The magnetoresistance was calculated by measuring the electrical resistivity in fields up to 50 kOe in the temperature range 2–300 K, using the linear four-probe technique. The resistivity was measured in the ac mode with a current of 100 mA and the excitation frequency was 33 Hz.

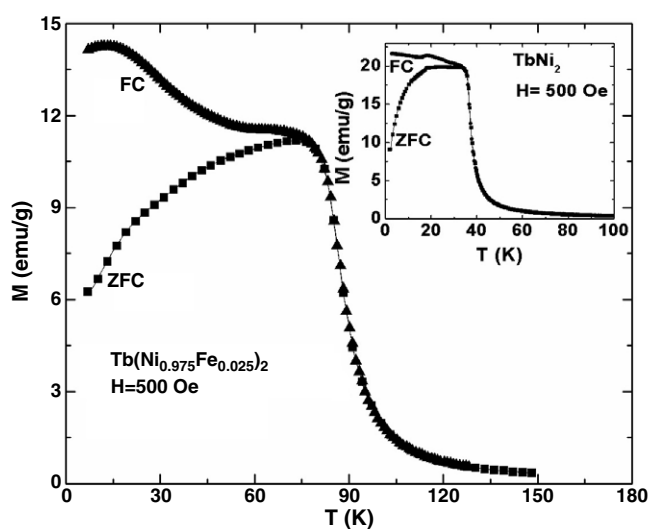


Figure 1. Temperature variation of ZFC and FC magnetization data of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$, obtained in an applied field of 500 Oe. The inset shows the M - T plot of TbNi_2 , realized under similar conditions.

3. Results and discussion

The Rietveld refinement of the room-temperature powder x-ray diffractograms confirmed that all the compounds crystallize in the MgCu_2 -type Laves phase cubic structure (space group $Fd\bar{3}m$, number 227). The refined lattice parameters (a) are 7.154 ± 0.001 , 7.183 ± 0.001 and 7.201 ± 0.001 Å, for the compounds with $x = 0$, 0.025 and 0.05, respectively. The temperature (T) dependence of magnetization (M) data, collected in an applied field (H) of 500 Oe, reveals that the ordering temperatures of the compounds with $x = 0$, 0.025 and 0.05 are 36 K, 87 K and 124 K, respectively. Figure 1 shows the M - T plot of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$ obtained in a field of 500 Oe, under zero-field-cooled (ZFC) and field-cooled (FC) conditions. A similar plot was obtained for $\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$ as well. The large thermo-magnetic irreversibility between ZFC and FC magnetization data seen in these compounds is attributed to the domain wall pinning effect [13]. Another feature worth noting in this figure is that the FC magnetization of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$, at low temperatures, shows a rather faster reduction with increase in temperature. This behaviour is found to be even more prominent in the case of $\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$. The inset of figure 1 shows the ZFC and FC magnetization plots of TbNi_2 . As can be seen from the inset, TbNi_2 shows an anomaly in the form of a kink at about 15 K, in both the FC and ZFC magnetization.

We have also studied the field dependence of magnetization data at various temperatures. Of particular interest are the M - H isotherms obtained at temperatures close to about half of the ordering temperature for all the compounds, and the representative plots for $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$ are given in figure 2. The M - H isotherms of TbNi_2 (at temperatures close to $T_C/2$) are given as an inset of figure 2. It may be noticed that the M - H isotherms of TbNi_2 at high fields show a better saturation tendency, compared to that of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$. It should be kept in mind that such a non-saturation could arise from the change in crystalline electric field (CEF) effects brought about by Fe substitution. However, the fact that the M - H isotherm of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$ at 4 K saturates in the same way as TbNi_2 may suggest that the CEF is not altered considerably by Fe substitution. Therefore, the above observations may be indicative

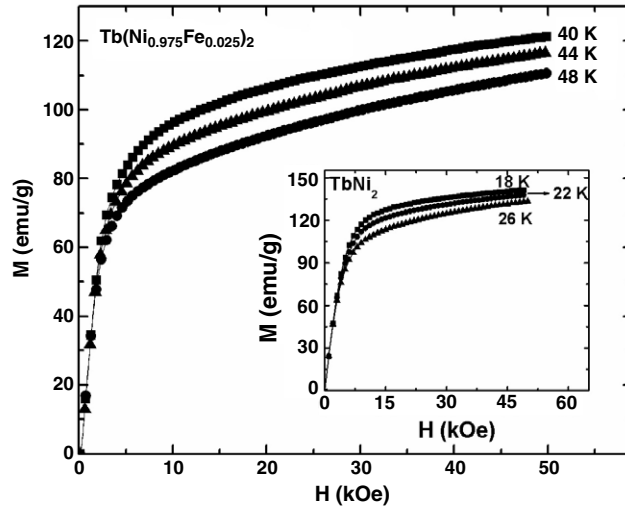


Figure 2. M - H isotherms of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$, obtained at about half of the ordering temperature. The inset shows the corresponding plots of TbNi_2 .

of the presence of a larger magnetic disorder in the Fe-substituted compounds as compared to that of TbNi_2 , in the same temperature range of T/T_C . The non-saturation feature seen in the Fe-substituted compounds could also arise due to the micro-inhomogeneities present in the sample. However, we rule out this possibility, as discussed at the end of this section.

To further probe the magnetic state of these compounds, we have carried out ac magnetic susceptibility (χ_{ac}) measurements with and without a dc bias field, in all these compounds. Figures 3(a) and (b) show the temperature variation of the real (χ'_{ac}) and the imaginary (χ''_{ac}) parts of the ac susceptibility normalized to the respective maximum values. It is clear from figure 3(a) that in all the compounds χ'_{ac} shows a peak at temperatures close to T_C . In the case of TbNi_2 , apart from the peak at T_C , another peak is observed at about 15 K, which is consistent with the observation seen in the M - T data. We attribute this peak to the spin reorientation transition reported in this compound [16]. Another interesting observation from these plots is the broadening of the χ'_{ac} peak in the Fe-substituted compounds. It is to be noted that though the FC magnetization data of the Fe-substituted compounds show some unusual reduction in the magnetization at low temperatures, no visible transition has been observed in the χ'_{ac} - T plot. However, the χ''_{ac} - T plots of these two compounds show a secondary peak at about 50 K.

In order to understand the nature of the low-temperature magnetic state, we have obtained the χ'_{ac} - T plot under a dc bias field of 20 kOe as well (figure 4). It can be seen that the sharpness of the peak corresponding to the spin reorientation transition (~ 15 K) for TbNi_2 has reduced considerably in the presence of the dc bias field. It has indeed been reported that a field of 10 kOe suppresses the spin reorientation transition in TbNi_2 [16]. Furthermore, one can see that the peak corresponding to the ordering temperature has shifted from 36 K to about 50 K, as the dc bias is applied. While the sharpness of the peak in TbNi_2 is marginally reduced with the dc bias, it has decreased considerably in the case of the Fe-substituted compounds. Furthermore, it is possible to note a small shoulder at about 25 K in the compound with $x = 0.025$, while a large peak appears in the compound with $x = 0.05$ at around 27 K. Based on the dc and ac magnetization data, it is not possible to attribute this peak to a spin reorientation transition.

Figure 5 shows the temperature variation of the heat capacity (C) for TbNi_2 in fields $H = 0$ and 50 kOe. The zero-field heat capacity data show a peak, which nearly coincides

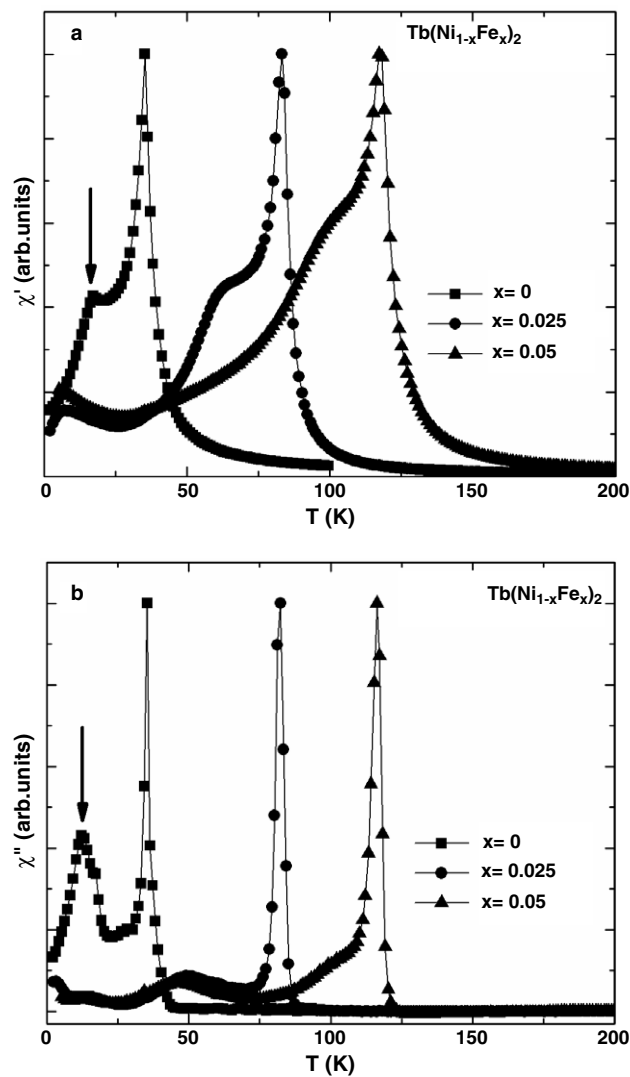


Figure 3. Temperature dependence of real (a) and imaginary (b) parts of normalized ac susceptibility of $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ compounds obtained under zero dc bias field. The frequency of measurement is 333 Hz. The arrows indicate the spin reorientation transitions seen in TbNi_2 .

with the T_C observed from the $M-T$ data. It may be noticed from the figure that, in the case of TbNi_2 , the peak associated with the magnetic transition in the $C-T$ plot starts at temperatures well above T_C . This may be attributed to the spin fluctuations, arising from the presence of magnetic polarons, above T_C . Such a behaviour has also been observed in other intermetallic compounds [17, 18]. However, in the iron-substituted compounds no peak in heat capacity could be seen even in zero field (inset in figure 5). Since the ordering temperatures of the iron-substituted compounds are considerably higher than that of TbNi_2 , the relatively larger lattice and electronic contributions to the total heat capacity presumably mask any weak peak due to the magnetic contribution. A similar behaviour has been observed in the $\text{Ho}(\text{Ni}, \text{Fe})_2$ system [15].

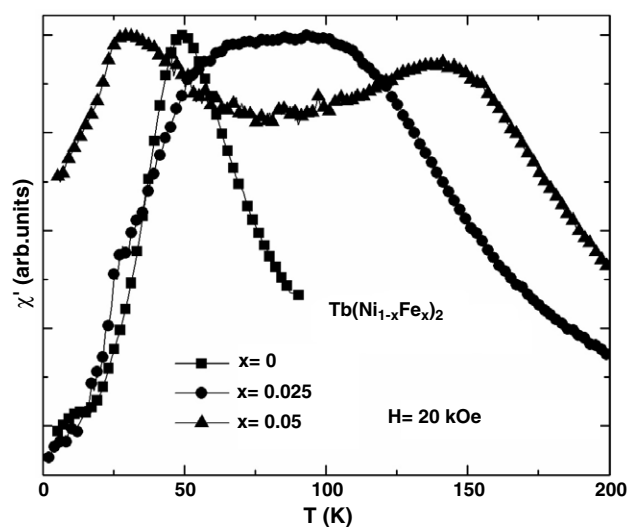


Figure 4. Temperature dependence of the real part of the normalized ac susceptibility of $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ compounds obtained under a dc bias field of 20 kOe. The frequency of measurement is 333 Hz.

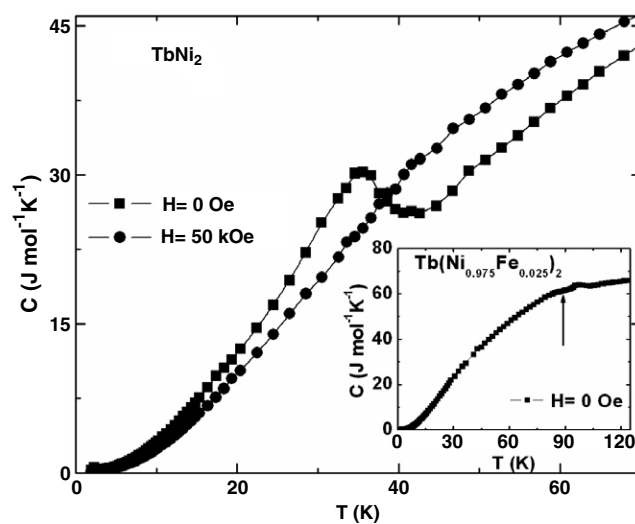


Figure 5. Heat capacity of TbNi_2 as a function of temperature in fields $H = 0$ and 50 kOe. The inset shows the variation of zero-field heat capacity in $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$. The arrow in the inset shows the ordering temperature determined from the magnetization data.

The magnetocaloric properties of $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ compounds, both from the magnetization data as well as from heat capacity data, have been determined using the methods reported earlier [15, 19]. Figure 6(a) shows the temperature variation of the isothermal magnetic entropy change (ΔS_M) using the $C-T$ data obtained in $H = 0$ and 50 kOe, for all the compounds. ΔS_M values calculated from the $C-H-T$ data were found to be in close agreement with those obtained from the $M-H-T$ data. It can be seen from the figure that the ΔS_M versus T plot for TbNi_2 , calculated for a field change (ΔH) of 50 kOe, shows a maximum

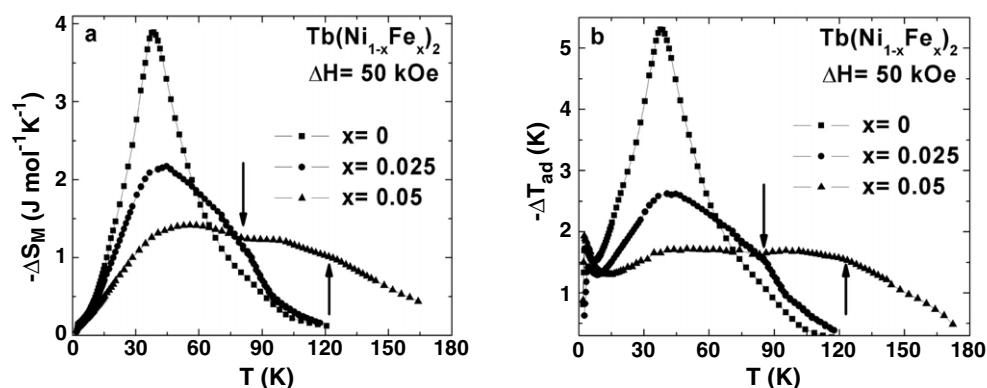


Figure 6. The temperature variation of magnetocaloric effect of $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ compounds, in terms of isothermal magnetic entropy change (a) and adiabatic temperature change (b), for a field change of 50 kOe. The arrows in the figure indicate the ordering temperatures.

near T_C with a value $4 \text{ J mol}^{-1} \text{ K}^{-1}$. The experimentally observed ΔS_M for TbNi_2 is less than the theoretically predicted [20] value of $\sim 7 \text{ J mol}^{-1} \text{ K}^{-1}$. The temperature variation of ΔS_M for $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$ show two broad peaks. Apart from a weak primary peak near T_C , a secondary peak at temperatures much below T_C is also observed. In the case of $\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$, a single broad peak with some signature of a small kink (close to T_C) is observed. The maximum value of ΔS_M (ΔS_M^{max}), for $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$, is found to be $\sim 2.2 \text{ J mol}^{-1} \text{ K}^{-1}$ at about 43 K, whereas for $\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$, it is $\sim 1.4 \text{ J mol}^{-1} \text{ K}^{-1}$ at about 53 K. The double-peak MCE behaviour seen here is quite similar to that reported in the $\text{Ho}(\text{Ni}, \text{Fe})_2$ system [15]. Figure 6(b) shows the temperature variation of the adiabatic temperature change (ΔT_{ad}), calculated from the $C-H-T$ data. Comparing figures 6(a) and (b), one can see that the temperature variation of ΔT_{ad} is identical to that of ΔS_M , except for a well-defined peak below 5 K, observed in all three compounds. Such a behaviour in the temperature variation of ΔT_{ad} has been reported in many other intermetallic compounds and may be attributed to the crystal field effect [21, 22].

The presence of a broad peak in the ΔS_M versus T plot of the iron-substituted compounds indicates the existence of some degree of magnetic randomness, at low temperatures, in these compounds. It may be recalled here that the $M-H$ isotherms of the Fe-substituted compounds, obtained at about half of the ordering temperature, indeed show some signature of a possible magnetic randomness at low temperatures (see figure 2). Relatively broad peaks seen in the $\chi_{\text{ac}}-T$ plots also indicate the possibility of randomness at low temperatures, in the case of Fe-substituted compounds. It is well known that the Ni sublattice in RNi_2 compounds is almost nonmagnetic [23] and hence the randomness cannot be attributed to Ni moments. Taking the maximum J value of $5/2$, as per Hund's rule, the theoretical magnetic entropy ($R \ln(2J + 1)$) associated with the Fe sublattice in the compounds with $x = 0.025$ and 0.05 is found to be 0.75 and $1.49 \text{ J mol}^{-1} \text{ K}^{-1}$ respectively. By comparing the theoretical values with the observed low-temperature MCE peak values, one can rule out the Fe contribution towards the low-temperature MCE peak. Therefore, the randomness occurring at low temperatures must be solely associated with the Tb sublattice. From a similar comparison of ΔS_M values associated with the primary peak with that of the theoretical magnetic entropy of the Fe sublattice, and taking into account the fact that in any magnetocaloric process only a fraction of the magnetic entropy is utilized [24], it can be inferred that ΔS_M values associated with the peak near T_C could not arise from the Fe sublattice alone. Therefore, it appears that the randomness starts at

low temperatures and continues up to T_C . This may be the reason for the considerable MCE in the region between the two peaks. We have earlier attributed a similar MCE behaviour observed in Fe-substituted HoNi_2 compounds to a possible randomization of Ho moments [15]. The randomization was thought to be a result of the change in the local anisotropy brought about by the Fe substitution.

The MCE behaviour of $\text{Tb}(\text{Ni}_{1-x}\text{Fe}_x)_2$ has also been studied for a field change of 10 kOe. The temperature variation of MCE of TbNi_2 , calculated for $\Delta H = 10$ kOe, is found to be similar to that obtained for 50 kOe, except for the peak values. However, in sharp contrast, in the Fe-substituted compounds the temperature variation of the MCE obtained for $\Delta H = 10$ kOe does not show the secondary peak. This indicates that a critical field (more than 10 kOe) is needed to bring about considerable ordering in the randomized Tb sublattice. Ideally, such an ordering is expected to give a metamagnetic-like transition in the $M-H$ isotherms at low temperatures. Though such a transition has indeed been observed in $\text{Ho}(\text{Ni}, \text{Fe})_2$ compounds [15], the $M-H$ isotherms obtained in the Fe-substituted TbNi_2 compounds do not show such a transition. Therefore, it seems that, in spite of the similarities in the MCE behaviour of $\text{Tb}(\text{Ni}, \text{Fe})_2$ and $\text{Ho}(\text{Ni}, \text{Fe})_2$ compounds, there is some difference in their magnetic structures at low temperatures.

Since a change in the magnetic state is expected to be reflected in the electrical resistivity behaviour, we have also studied the electrical resistivity of these compounds, both as a function of temperature and applied magnetic field. The temperature variation of the electrical resistivity of all the compounds, normalized to the value at 300 K, is shown in figure 7. It can be seen from the figure that all the compounds show metallic character. The onset of magnetic ordering in TbNi_2 is characterized by an anomaly in the resistivity, whereas in the Fe-substituted compounds a change in the slope of the normalized resistivity marks the ordering temperatures. It may be noticed from figure 7 that the resistivity of TbNi_2 , near the ordering temperature, initially shows an upturn and then shows a decrease on cooling. Such a behaviour is generally expected in antiferromagnetic materials [25]. However, in TbNi_2 , this behaviour of the resistivity may be attributed to the presence of the spin fluctuations. Partial polarization of the Ni 3d band resulting in the formation of non-zero magnetic moment on Ni may be mainly responsible for the enhancement of spin fluctuations. Just below T_C , the long-range magnetic order sets in, which suppresses the fluctuations, thereby causing a drop in the resistivity. It may be recalled here that the temperature variation of heat capacity data of TbNi_2 is also indicative of the presence of spin fluctuations just above T_C . Such a resistivity behaviour has been observed in other intermetallic compounds as well [18].

The field dependence of magnetoresistance (MR), defined as $(\frac{R(H)-R(0)}{R(0)})$, for all the compounds has been studied in various temperature regions. The MR isotherms of TbNi_2 and $\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$, above their ordering temperatures are shown in figure 8. It can be seen from the figure that near T_C , the MR value for TbNi_2 is negative; its magnitude decreases with increasing temperature and finally becomes positive at higher temperatures. This observation is in contrast with the behaviour seen in HoNi_2 , which showed a positive MR at temperatures close to T_C [15]. The positive MR in HoNi_2 was attributed to the dominant contribution from the Lorentz force term [26]. Since the Lorentz force contribution in both these compounds is expected to be almost equal, the presence of negative MR in TbNi_2 close to T_C indicates that there is a large negative contribution in this case. The difference between the MR behaviours of HoNi_2 and TbNi_2 may be due to the difference in the exchange coupling strengths in these two compounds. It is to be noted that the T_C values are 14 K and 36 K for HoNi_2 and TbNi_2 , respectively.

In the Fe-substituted compounds, on the other hand, the MR near T_C is strongly negative. While the maximum value of the MR for TbNi_2 is about 1.6%, it is about 6.7% for

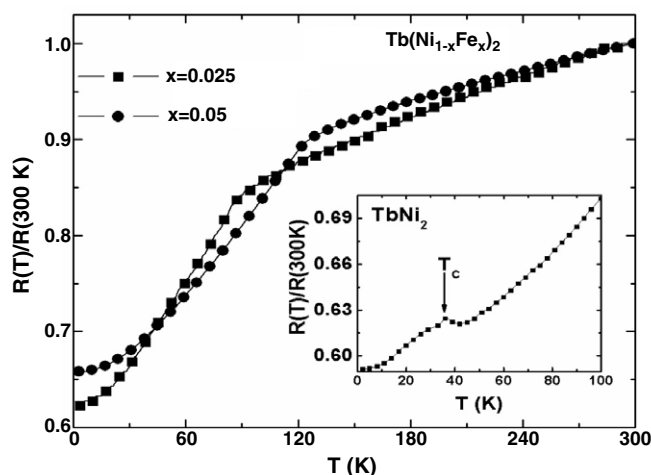


Figure 7. Temperature variation of electrical resistivity, normalized to the value at 300 K, of $\text{Tb}(\text{Ni}, \text{Fe})_2$ compounds. The inset shows the temperature dependence of normalized resistivity of TbNi_2 . The arrow in the inset shows the T_C of TbNi_2 .

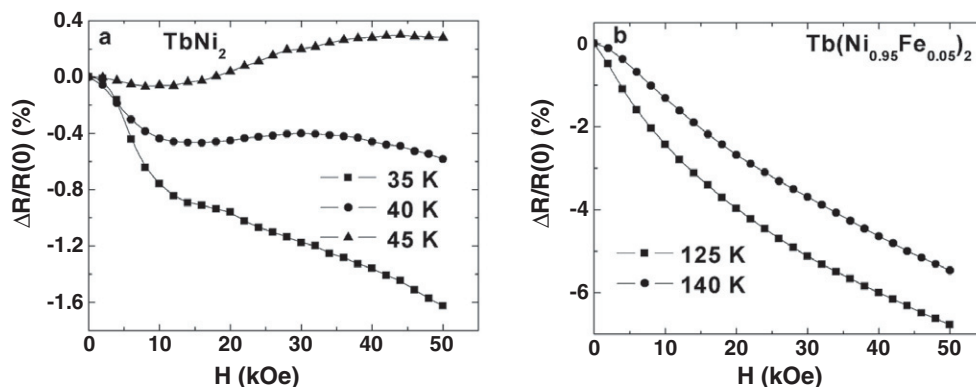


Figure 8. Field dependence of MR in TbNi_2 (a) and $\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$ (b) compounds, near their ordering temperatures.

$\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$, for a field of 50 kOe. The maximum value of the MR for $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$ is found to be $\sim 8.5\%$ for the same field. The presence of large MR in the Fe-substituted compounds may be attributed to suppression of spin fluctuations. It is well known that the MR in many R–TM systems originates from the suppression of spin fluctuations.

Figure 9 shows the temperature variation of the electrical resistance of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$, obtained under various applied magnetic fields. Application of magnetic field reduces the resistance, rendering the MR negative. The suppression of the resistance with the application of the field is maximum near the ordering temperature. It may be noticed that the suppression of the electrical resistance, with the application of the 50 kOe field, starts at about 160 K. This indicates the presence of an additional magnetic contribution to the electrical resistance, in the Fe-substituted compounds, at temperatures well above T_C . We attribute this to the presence of spin fluctuations, as mentioned earlier. A similar behaviour has been reported in other intermetallic compounds as well [18]. The fact that the MR at 10 kOe is appreciable only in the

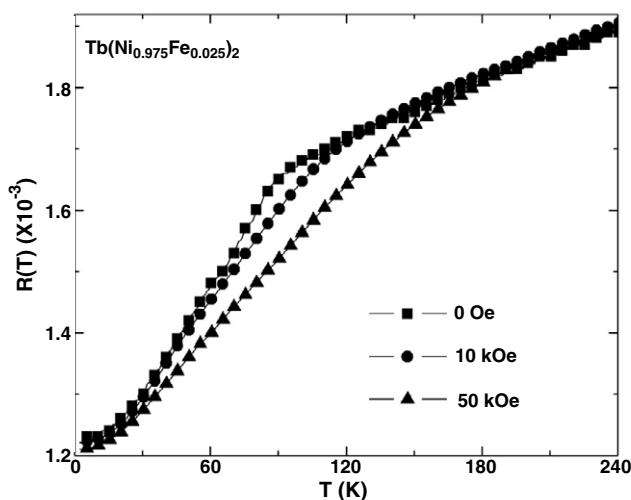


Figure 9. Temperature variation of electrical resistance of $\text{Tb}(\text{Ni}_{0.975}\text{Fe}_{0.025})_2$, obtained under various applied magnetic fields.

vicinity of T_C may be due to the competing effect from the positive Lorentz force contribution and the negative spin fluctuation contribution.

In order to throw more light on the low-temperature behaviour of the Fe-substituted compounds, especially near the temperature corresponding to the secondary MCE peak, the field dependence of the MR has been estimated, and the representative plot of $\text{Tb}(\text{Ni}_{0.95}\text{Fe}_{0.05})_2$ is shown in figure 10. It can be seen from the figure that, in this temperature range, the MR is almost insensitive to the field up to 10 kOe, but it increases considerably at higher fields. Since a negative MR could arise due to the suppression of the excess resistivity by the applied field, it is reasonable to assume that some amount of magnetic disorder is present under zero-field conditions in these compounds, at low temperatures. At this point, it is of importance to recall the signature of randomization seen in the magnetic (dc and ac) and the magnetocaloric results in the case of Fe-substituted compounds.

Therefore, we find that the magnetic, magnetocaloric and magnetoresistance behaviour in the Fe-substituted TbNi_2 compounds shows considerable similarities to that of $\text{Ho}(\text{Ni}, \text{Fe})_2$. The anomalous behaviour seen in both these systems seems to arise from the unusual magnetic randomness occurring in the rare earth sublattice at low temperatures. Different experimental findings seem to support the proposition of randomization of the R moments. Since the TM sublattice is inherently nonmagnetic, RKKY interaction, which is generally weak, is the only exchange mechanism present in these compounds. Upon Fe substitution, there may be a modulation of the R–R exchange, resulting in the fanning of R moments. Due to the cubic structure, the magneto-crystalline anisotropy would be small, and therefore, the fanned R moments take random orientations. This would cause an increase in the magnetic entropy at low temperature. On application of a field, these random moments align and the entropy decreases. This may be the reason for the unexpected positive MCE and negative MR behaviour in the Fe-substituted compounds. It may be mentioned here that secondary phases and micro-inhomogeneities could also give rise to a similar anomalous behaviour. To rule out this possibility, we have carried out microstructural studies, which clearly show that the samples are indeed homogeneous and single phase. In view of this, a detailed neutron diffraction study would be essential to probe the exact magnetic structure, especially at low temperatures.

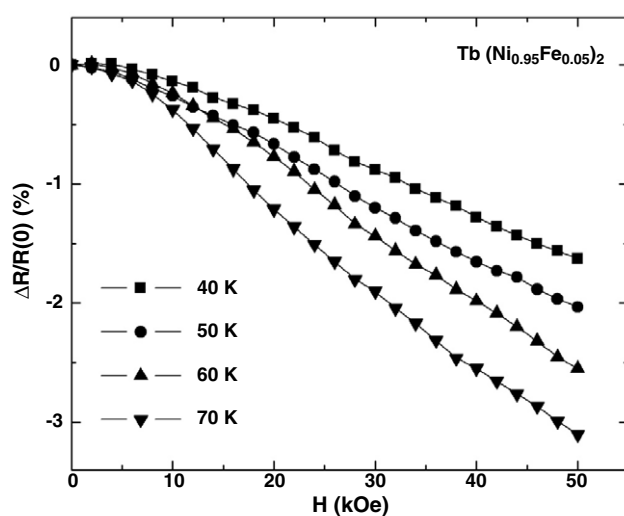


Figure 10. Field dependence of MR in Tb(Ni_{0.975}Fe_{0.025})₂ at low temperatures.

4. Conclusions

In conclusion, the present study shows that MCE and MR show some anomalous behaviour in the iron-substituted TbNi₂ compounds. It seems that the partial substitution of Fe for Ni leads to the randomization of Tb moments at low temperatures, which gives rise to a considerable MCE over a wide range of temperature. We feel that the randomization of the Tb sublattice is due to the local anisotropy variations caused by the partial substitution of magnetic Fe in place of nonmagnetic Ni. The present study also shows that the anomalous magnetocaloric and magnetoresistance behaviour seen in the present case is similar to that obtained in the Fe-substituted HoNi₂ compounds.

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