A comparative study of magnetic behaviors in TbNi2, TbMn2 and TbNi2Mn

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A comparative study of magnetic behaviors in TbNi2, TbMn2 and TbNi2Mn

Abstract
All TbNi2, TbMn2, and TbNi2Mn compounds exhibit the cubic Laves phase with AB2-type structure in spite of the fact that the ratio of the Tb to transition-metal components in TbNi2Mn is 1:3. Rietveld refinement indicates that in TbNi2Mn the Mn atoms are distributed on both the A (8a) and B (16d) sites. The values of the lattice constants were measured to be a = 14.348 Å (space group F-43 m), 7.618 Å, and 7.158 Å (space group Fd-3 m) for TbNi2, TbMn2, and TbNi2Mn, respectively. The magnetic transition temperatures TC were found to be TC = 38 K and TC = 148 K for TbNi2 and TbNi2Mn, respectively, while two magnetic phase transitions are detected for TbMn2 at T1 = 20 K and T2 = 49 K. Clear magnetic history effects in a low magnetic field are observed in TbMn2 and TbNi2Mn. The magnetic entropy changes have been obtained.

Keywords
tbni2, tbmn2, tbni2mn, comparative, study, magnetic, behaviors

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A comparative study of magnetic behaviors in TbNi$_2$, TbMn$_2$ and TbNi$_2$Mn


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All TbNi$_2$, TbMn$_2$, and TbNi$_2$Mn compounds exhibit the cubic Laves phase with AB$_2$-type structure in spite of the fact that the ratio of the Tb to transition-metal components in TbNi$_2$Mn is 1:3. Rietveld refinement indicates that in TbNi$_2$Mn the Mn atoms are distributed on both the A (8a) and B (16d) sites. The values of the lattice constants were measured to be $a = 14.348 \text{ Å}$ (space group F$-43 m$), $7.618 \text{ Å}$, and $7.158 \text{ Å}$ (space group Fd$-3 m$) for TbNi$_2$, TbMn$_2$, and TbNi$_2$Mn, respectively. The magnetic transition temperatures $T_C$ were found to be $T_C = 38 \text{ K}$ and $T_C = 148 \text{ K}$ for TbNi$_2$ and TbNi$_2$Mn, respectively, while two magnetic phase transitions are detected for TbMn$_2$ at $T_1 = 20 \text{ K}$ and $T_2 = 49 \text{ K}$. Clear magnetic history effects in a low magnetic field are observed in TbMn$_2$ and TbNi$_2$Mn. The magnetic entropy changes have been obtained.

Rare earth (R) compounds with RM$_2$ Laves phase structure offer the unique opportunity to investigate systematically the influence of occupation and localization of f and d orbitals on the electronic and magnetic properties of solids. The RNi$_2$ compounds are often considered to show the cubic Laves phase structure ($C_{15}$-Fd$-3 m$ space group) but recent studies indicated that RNi$_2$ compounds crystallize in a superstructure of $C_{15}$ with ordered vacancies at the R sites (with F$-43 m$ space group) and external pressure or temperature can induce reversible disordering of the R vacancies in the RNi$_2$ Laves phase superstructures (transformation from F$-43 m$ to Fd$-3 m$). RM$_2$ system crystallizes in cubic Laves phase structure ($C_{15}$ type) or hexagonal Laves phase structure ($C_{14}$ type) depending on the rare earth element and annealing temperature. TbMn$_2$ locates at a critical condition for the collapse of Mn moments where the Mn moment is highly unstable and it is easily collapsed by applying pressure or even a magnetic field. Recently, we reported that, similar to RNi$_2$ alloys, the RN$_2$Mn with $R = \text{Tb, Dy, Ho, Er}$ alloys also crystallize in the cubic MgCu$_2$-type of structure with the Mn atoms occupying both the R and the Ni lattice positions.

Here, we present a comparative study of TbNi$_2$, TbMn$_2$, and TbNi$_2$Mn compounds to get a deeper understanding of the structural and magnetic properties of these compounds. TbNi$_2$Mn$_{1.0}$, Tb$_{1-x}$Ni$_2$ ($x = 0$, 0.02, and 0.05), and TbMn$_2$ alloys were prepared by arc-melting procedures. All samples were characterized by x-ray diffraction (CuK$_\alpha$ radiation; $\lambda = 1.5418 \text{ Å}$). Magnetization measurements were carried out in a conventional physical properties measurement system. The neutron diffraction patterns of the TbNi$_2$Mn sample were collected at the Wombat, ANSTO ($\lambda = 2.4072 \text{ Å}$) over 5–300 K.

Our x-ray diffraction results (Figure 1) at room temperature indicate that, among the Tb$_{1-x}$Ni$_2$ ($x = 0$, 0.02, and 0.05) compounds, only the sample with $x = 0.02$ is single phase and other compositions samples include larger amount of impurities such as TbNi and TbNi$_3$ phase. This agrees well with Ref. 2, where it was reported that single phase samples with superstructure of the cubic RNi$_2$ Laves phase

![Figure 1](image_url)

**Figure 1.** Room temperature X-ray diffraction patterns with refinement results.
can only be obtained in a R deficient composition and the accurate composition (the ratio of Tb: Ni) in a good TbNi$_2$ sample is determined to be around 0.98:2. Our refinements demonstrate that the Tb$_{0.98}$Ni$_2$ compound crystallizes in the superstructure of the cubic RNi$_2$ Laves phase (F-43m space group) with the doubled lattice parameter of C15 being a = 14.348(1) Å, which is close to 14.342 Å (at 295 K) reported in Ref. 2, while both TbNi$_2$Mn and TbMn$_2$ exhibit the cubic Laves phase structure with space group Fd-3m. For simplicity, we use TbNi$_2$ to represent our Tb$_{0.98}$Ni$_2$ sample, a field of 0.01 T on warming process after first cooling in zero field. Figure 2 shows the temperature dependence of DC magnetization for three samples. Both TbNi$_2$ and TbNi$_2$Mn exhibit standard ferromagnetic behavior with Tc values being derived to be 38 K for TbNi$_2$ and 148 K for TbNi$_2$Mn, respectively. On the other hand, the TbMn$_2$ compound shows two magnetic phase transitions: one at $T_1 = 20$ K and another at $T_2 = 49$ K, respectively, which agrees well with previous report.

It can be seen that different thermo-magnetic behavior is detected for the ZFC and FC magnetization curves for TbNi$_2$Mn and TbMn$_2$ compounds. The presence of magneto-history effects in these two samples may be ascribed to the presence of narrow Bloch walls.

In other rare earth–Mn based compounds where obvious magneto-history effects were detected, such as R$_2$Fe$_{17-x}$Mn$_x$ (with R = Nd, Pr, and Er) and R$_2$Fe$_{14-x}$Mn$_x$C (with R = Pr and Ho), it was considered that the fluctuation of the crystal fields as well as the exchange fields in the local environments due to the introduction of Mn atoms are responsible for the magneto-history effects detected there. In the present case, it is reasonable to assume that the Mn atoms in both TbNi$_2$Mn and TbMn$_2$ may also lead to similar fluctuation and contribute to the observed magneto-history effects (Figure 2). Moreover, as shown by the inset in Figure 2, above $T_C$ the inverse susceptibility of TbNi$_2$ follows Curie-Weiss behaviour leading to a paramagnetic Weiss temperature $\theta$ of 37.8 K, and an effective magnetic moment of $\mu_{eff} = 7.0 \mu_B$.

We have measured the $M^{-\mu_H}$ curves around the transition temperatures and obtained the corresponding Arrott-plots of $M^2$ versus $\mu_H$/$M$. The data (not shown here) for TbNi$_2$ and TbNi$_2$Mn (Ref. 6) indicate that the phase transitions around $T_C$ in these two compounds are both second order while different magnetic behaviors are observed at $T_1 = 20$ K and $T_2 = 49$ K for TbMn$_2$. The field dependences of magnetization for TbMn$_2$ in the regions of $T_1$ (below 35 K) and $T_2$ (between 40 K and 65 K) are drawn in Figures 3(a) and 3(c).
respectively. It can be clearly seen that around $T_2$ the $M-\mu_0H$ curves for increasing and decreasing fields are the same while around $T_1$ the $M-\mu_0H$ curves are significantly different. This indicates that the transition at $T_2$ is second order while the transition at $T_1$ is first order. The nature of the transitions at $T_2$ (second order) and $T_1$ (first order) is drawn out more clearly by the corresponding Arrott-plots (Figures 3(d) and 3(b)), respectively.

The difference in the order of the magnetic phase transitions at $T_1$ and $T_2$ is supported by previous thermal expansion results. It was reported that TbMn$_2$ exhibits a volume expansion of 1.68% accompanied by a monoclinic distortion around $T_1$ while the magneto-volume effect is much less clear around $T_2$. On the other hand, it was reported that the thermal expansion of TbNi$_2$ exhibits a slight anomaly (spontaneous volume strain $\sim 6 \times 10^{-5}$) around $T_C$. Moreover, it was reported that $dT_C/dP$ is equal to 1.96 K/Gpa for TbNi$_2$Mn (Ref. 13) while by comparison, the values in TbMn$_2$ are $dT_C/dP = 2$ K/kilobars at the $T_2$ transition and $dT_C/dP = -30$ K/kilobars at $T_1$. Furthermore, it should be noted that the magnetization curves exhibit a large high field susceptibility in TbMn$_2$ (see Figures 3(a) and 3(c)), which may reflect the possibility of the existence of non-collinear magnetic moment structure in this compound.

The magnetic entropy changes around the magnetic transition temperatures have been derived from the magnetization curves based on the Maxwell thermodynamic relation. For a field change from 0 T to 2 T, the maximum of $-\Delta S_M$ value is derived to be 4.8 J kg$^{-1}$ K$^{-1}$ (around 41 K) for TbNi$_2$, 1.4 J kg$^{-1}$ K$^{-1}$ (around 136 K) for TbNi$_2$Mn, and 4.7 J kg$^{-1}$ K$^{-1}$ (around 51 K) for TbMn$_2$, respectively.

We have also measured the ac susceptibility components of the TbMn$_2$ (Figure 4(a)) and TbNi$_2$Mn (Figure 4(b)) compounds. It is accepted that for highly anisotropic materials, the value of $\gamma'$ is determined mainly by the magnetic anisotropy energy and the domain-wall energy, whereas the value of $\gamma''$ gives the energy absorption.

Figure 4(a) indicates clearly the presence of two transitions (marked by arrows) in TbMn$_2$. The magnetic phase transition in TbNi$_2$Mn compound can also be detected clearly by the ac susceptibility measurements of Figure 4(b) with evidence for an additional anomaly at lower temperatures around 25 K in the $\chi''$ versus $T$ curve. This latter anomaly is likely to be related to magnetic domain movement. The magnetic phase transition around $T_C = 148$ K in TbNi$_2$Mn has also been confirmed by neutron diffraction measurements over the temperature range of 10–300 K. The temperature dependences of the intensities of the (111) and (220), are shown in the inset of Figure 4(b). The increase in peak intensity with decreasing temperature reflects clearly the onset of the ferromagnetic phase transition around $T_C \sim 150$ K, in good agreement with the magnetic data.

Application of a DC applied field during the ac susceptibility measurement process can eliminate the ac susceptibility response to technical magnetization effects such as domain walls while suppressing the background. In addition, an applied DC field ensures that only critical data are analysed [Ref. 6 and references therein]. This allows the critical exponents that characterize the nature of the divergence to be derived as functions of temperature and DC applied field [Ref. 6 and references therein]. As is evident from Figure 4(a), the magnitude of the ac susceptibility peak around $T_2$ decreases and the peak position shifts to higher temperature with increasing DC field, consistent with the behaviour expected for a second order transition [Ref. 6 and reference therein]. Moreover, we also find the peak position is found to shift slightly towards higher temperature with a change of frequent from $f = 50$ Hz to 3200 Hz (not shown here), which may reflect the possible presence of spin glass-like behaviour in this compound.

![Figure 4](image-url)

**FIG. 4.** (a) The ac magnetic susceptibility of TbMn$_2$ as measured at the dc fields listed. Upper panel: real part, $\chi'$; lower panel: Imaginary part, $\chi''$. (b) The ac magnetic susceptibility of TbNi$_2$Mn (ac magnetic field $\mu_0H_{ac} = 0.0004$ T; $f = 1000$ Hz; $\mu_0H_{dc} = 0.01$ T). Upper panel: real part, $\chi'$; lower panel: Imaginary part, $\chi''$. The inset shows the temperature dependences of the intensities of the two main magnetic peaks, (111) and (220), as determined from neutron diffraction studies of TbNi$_2$Mn.