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

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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

Disciplines

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A comparative study of magnetic behaviors in $TbNi_2$, $TbMn_2$ and $TbNi_2Mn$

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All $TbNi_2$, $TbMn_2$, and $TbNi_2Mn$ 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 $TbNi_2Mn$ is 1:3. Rietveld refinement indicates that in $TbNi_2Mn$ 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 $TbNi_2$, $TbMn_2$, and $TbNi_2Mn$, respectively. The magnetic transition temperatures T_C were found to be $T_C = 38$ K and $T_C = 148$ K for $TbNi_2$ and $TbNi_2Mn$, respectively, while two magnetic phase transitions are detected for $TbMn_2$ at $T_1 = 20$ K and $T_2 = 49$ K. Clear magnetic history effects in a low magnetic field are observed in $TbMn_2$ and $TbNi_2Mn$. The magnetic entropy changes have been obtained. © *2014 AIP Publishing LLC*.

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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.[1] 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*).[2,3] RMn_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.[4] $TbMn_2$ 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[5,6] reported that, similar to RNi_2 alloys, the RNi_2Mn with R = 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.[5]

Here, we present a comparative study of $TbNi_2$, $TbMn_2$, and $TbNi_2Mn$ compounds to get a deeper understanding of the structural and magnetic properties of these compounds.

$TbNi_2Mn_{1.0}$, $Tb_{1-\delta}Ni_2$ ($\delta = 0$, 0.02, and 0.05), and $TbMn_2$ alloys were prepared by arc-melting procedures. All samples were characterized by x-ray diffraction (CuKα radiation; $\lambda = 1.5418$ Å). Magnetization measurements were carried out in a conventional physical properties measurement system. The neutron diffraction patterns of the $TbNi_2Mn$ sample were collected at the Wombat, ANSTO ($\lambda = 2.4072$ Å) over 5–300 K.

Our x-ray diffraction results (Figure 1) at room temperature indicate that, among the $Tb_{1-\delta}Ni_2$ ($\delta = 0$, 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

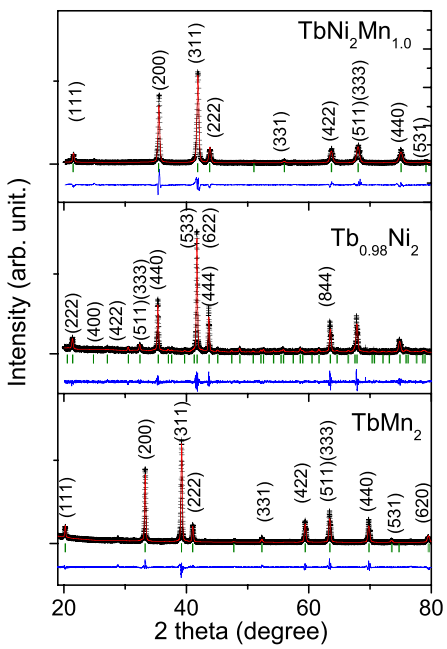

FIG. 1. Room temperature X-ray diffraction patterns with refinement results.

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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-43 m 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 $TbNi_2Mn$ and $TbMn_2$ exhibit the cubic Laves phase structure with space group Fd-3 m. For simplicity, we use $TbNi_2$ to represent our $Tb_{0.98}Ni_2$ sample in this manuscript. It is worth noting that the lattice constant of $TbNi_2Mn$ (a = 7.158(1) Å) is much closer to that of $TbNi_2$ (we use half of lattice constant in $TbNi_2$ for comparison ~7.174 Å; this is based on the fact that F-43 m is a superstructure of C15 with double the lattice parameter of C15) rather than the $TbMn_2$ (a = 7.618(1) Å) which is almost 6.5% larger than the lattice parameter of $TbNi_2Mn$. This indicates that the bonding distances between atoms in $TbMn_2$ are significantly larger than in $TbNi_2Mn$ and $TbNi_2$. In the $TbNi_2Mn$ compound, the Mn atoms are found to be distributed on both the A (8a site) and B (16d) sites.[5] It is also noted that a recent report shows that $TbNi_2$ with a pure C15 phase can also be obtained with lattice constant a = 7.172(4),[7] which is very close of our value (half of a for F-43 m) mentioned above for $TbNi_2$.

The samples have been measured in an applied field of $\mu_0H = 0.01$ T on warming process after first cooling in zero fields (marked as ZFC). The field-cooled cooling magnetization curve, FC, was then recorded on cooling in $\mu_0H = 0.01$ T from 300 K to 5 K after ZFC. For the $TbNi_2Mn$ sample, a field of $\mu_0H = 0.005$ T was used. Figure 2 shows the temperature dependence of DC magnetization for three samples. Both $TbNi_2$ and $TbNi_2Mn$ exhibit standard ferromagnetic behavior with T_C values being derived to be 38 K for $TbNi_2$ and 148 K for $TbNi_2Mn$, 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.[8] It can be seen that different thermo-magnetic behavior is detected for the ZFC and FC magnetization curves for $TbNi_2Mn$ and $TbMn_2$ compounds. The presence of magneto-history effects in these two samples may be ascribed to the presence of narrow Bloch walls.[5,9] In other rare earth–Mn based compounds where obvious magneto-history effects were detected, such as $R_2Fe_{17-x}Mn_x$ (with R = Nd, Pr, and Er) and $R_2Fe_{14-x}Mn_xC$ (with R = Pr and Ho),[10] 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.[10] In the present case, it is reasonable to assume that the Mn atoms in both $TbNi_2Mn$ 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 $\theta_p = 37.8$ K, and an effective magnetic moment of $\mu_{eff} = 7.0\,\mu_B$.

We have measured the M-μ_0H curves around the transition temperatures and obtained the corresponding Arrott-plots of M^2 versus μ_0H/M. The data (not shown here) for $TbNi_2$ and $TbNi_2Mn$ (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),

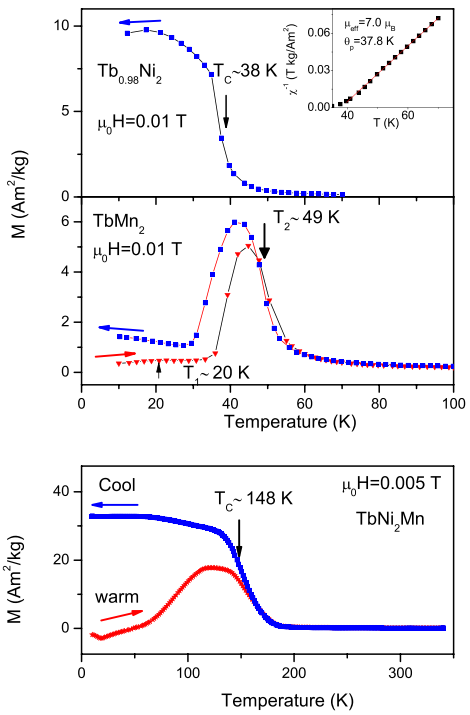

FIG. 2. Temperature dependencies of the dc magnetization of $TbNi_2$, $TbMn_2$, and $TbNi_2Mn$ measured after first cooling in zero field. Note the strong magneto-history effect in $TbMn_2$ and $TbNi_2Mn$. The inset shows the temperature dependence of the inverse susceptibility for $TbNi_2$.

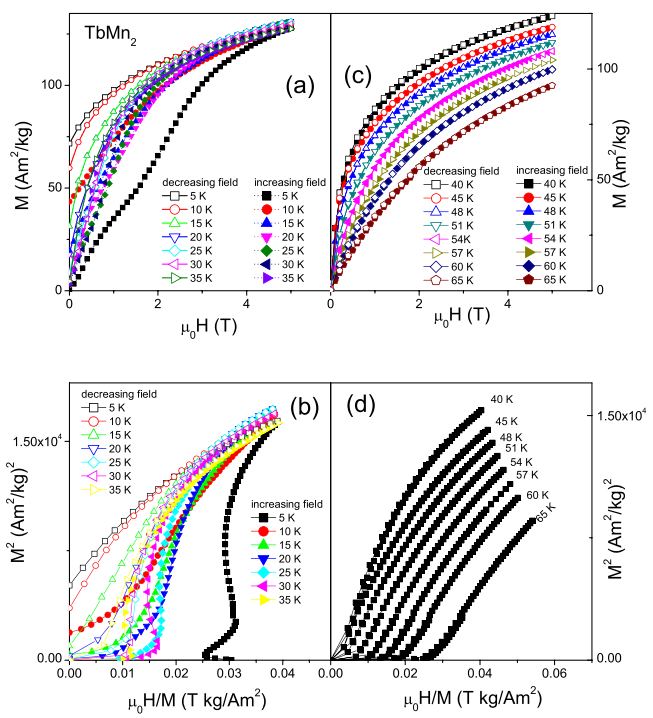

FIG. 3. (a) and (c) The magnetization isotherms of $TbMn_2$ over 5–35 K ($T_1 = 20$ K) and 40–65 K ($T_2 = 49$ K), respectively. Figs. 3(b) and 3(d) show the corresponding Arrott curves of M^2 versus μ_0H/M.

respectively. It can be clearly seen that around T_2 the M-μ_0H curves for increasing and decreasing fields are the same while around T_1 the M-μ_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.[11] 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.[11] 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.[12] Moreover, it was reported that dT_C/dP is equal to 1.96 K/Gpa for $TbNi_2Mn$ (Ref. 13) while by comparison, the values in $TbMn_2$ are $dT_C/dp = 2$ K/kilobars at the T_2 transition and $dT_N/dp = -30$ K/kilobars at T_1.[14] 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.[15]

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 $TbNi_2Mn$, 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 $TbNi_2Mn$ (Figure 4(b)) compounds. It is accepted that for highly anisotropic materials, the value of χ' is determined mainly by the magnetic anisotropy energy and the domain-wall energy, whereas the value of χ'' gives the energy absorption.[16]

Figure 4(a) indicates clearly the presence of two transitions (marked by arrows) in $TbMn_2$. The magnetic phase transition in $TbNi_2Mn$ compound can also be detected clearly by the ac susceptibly measurements of Figure 4(b) with evidence for an additional anomaly at lower temperatures around 25 K in the χ'' 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 $TbNi_2Mn$ 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

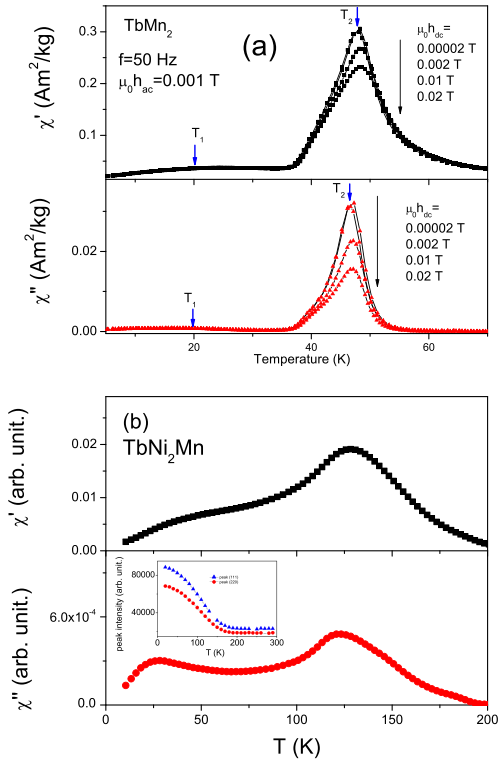

FIG. 4. (a) The ac magnetic susceptibility of $TbMn_2$ as measured at the dc fields listed. Upper panel: real part, χ'; lower panel: Imaginary part, χ''. (b) The ac magnetic susceptibility of $TbNi_2Mn$ (ac magnetic field $\mu_0 h_{ac} = 0.0004$ T; $f = 1000$ Hz; $\mu_0 h_{dc} = 0.01$ T). Upper panel: real part, χ'; lower panel: Imaginary part, χ''. 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 $TbNi_2Mn$.

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),[17] which may reflect the possible presence of spin glass-like behaviour in this compound.

[1]K. H. J. Buschow, Rep. Prog. Phys. **40**, 1179 (1977).
[2]E. Gratz *et al.*, J. Phys.: Condens. Matter **11**, 7893 (1999).
[3]A. Lindbaum *et al.*, Phys. Rev. B **65**, 134114 (2002).
[4]Y. Makihara *et al.*, J. Phys. Soc. Jpn. **52**, 629 (1983).
[5]J. L. Wang *et al.*, Phys. Rev. B **73**, 094436 (2006).
[6]J. L. Wang *et al.*, J. Phys.: Condens. Matter **23**, 216002 (2011).
[7]S. Delsante *et al.*, J. Chem. Thermodyn. **65**, 73 (2013).
[8]S. Labroo *et al.*, J. Appl. Phys. **67**, 5292 (1990).
[9]J. L. Wang *et al.*, Solid State Commun. **121**, 615 (2002).
[10]Z. G. Sun *et al.*, J. Alloys Compd. **349**, 1 (2003).
[11]P. J. Brown *et al.*, J. Phys.: Condens. Matter **4**, 1103 (1992); M. Shiga, Physica B **149**, 293 (1988).
[12]M. R. barra *et al.*, J. Phys. Chem. Solids **45**, 789 (1984).
[13]D. D. Jackson *et al.*, Phys. Rev. B **75**, 224422 (2007).
[14]M. R. Ibarra *et al.*, J. Appl. Phys. **75**, 7158 (1994).
[15]J. L. Wang *et al.*, J. Appl. Phys. **91**, 2165 (2002).
[16]J. L. Wang *et al.*, Phys. Rev. B **67**, 014417 (2003).
[17]F. Hong *et al.*, "$TbMn_2$: The simplest compound with the exchange bias effect" (submitted).