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Magnetic phase transitions in (Tb,Y)Mn_2M_2 (M=Ge and Si) systems

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The temperature- and field-induced magnetic phase transitions in pseudoternary systems $Tb_{1-x}Y_xMn_2Ge_2$ and $Tb_{1-x}Y_xMn_2Si_2$, which are characterized by a separate magnetic ordering of the Mn and Tb subsystems, have been studied on strongly textured samples by magnetization and thermal expansion measurements. In the concentration region $0.4 \leqslant x \leqslant 1.0$, where the Mn moments are ordered only, the magnetic structure is antiferromagnetic. The magnetic ordering in the Tb subsystem is accompanied by a transformation of the Mn magnetic structure. As a result, the compounds show a spontaneous magnetization. This transition is of a first-order type. In both systems the transition temperature for the Tb subsystem $T_C^{(Tb)}$ decreases linearly with increasing Y concentration and goes to zero at about $x=0.40$. Canted phases and field-induced first-order transitions into the collinear phase have been observed in both systems below this temperature. It is concluded that the $f-d$ exchange interaction is the primary factor governing the value of $T_C^{(Tb)}$.
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INTRODUCTION

The ternary intermetallic compounds RMn_2X_2 with X = Ge or Si are antiferromagnets at high temperature ($T_N > 400$ K) and attract interest due a number of temperature-induced magnetic phase transitions. These compounds crystallize in the $ThCr_2Si_2$-type tetragonal structure formed by rare earth (RE), Mn, and X layers in the sequence R–X–Mn–X–R perpendicular to the tetragonal axis. One of the interesting properties of the RMn_2X_2 compounds with magnetic RE is that the RE sublattice remains in the paramagnetic state below T_N and orders at $T_C^{(R)} \sim 100$ K.[1] In the temperature interval $T_C^{(R)} < T < T_N$, the magnetic structure of the Mn sublattice depends on the in-plane Mn–Mn distance. When this distance is smaller than about 2.86 Å a spontaneous magnetization appears in the Mn sublattice.[2,3] Below $T_C^{(R)}$ various collinear and noncollinear ferrimagnetic structures are stabilized in these compounds.[4–7]

The $TbMn_2X_2$ compounds show a relatively simple behavior. These compounds have been studied by different methods.[4,6–8,9] However, the results obtained are controversial. According to recent data,[4] $TbMn_2Ge_2$ has a collinear ferrimagnetic structure at $T < T_C^{(Tb)}$, which transforms into a canted one at $T_{tr} \sim 30$ K. $T_C^{(R)}$ varies from 95 to 110 K.[1] In $TbMn_2Si_2$, the negatively coupled Tb and Mn sublattices form a collinear ferrimagnetic structure below $T_C^{(Tb)}$ as reported in Ref. 9. However, following Ref. 6 in $TbMn_2Si_2$ a collinear antiferromagnetic structure of the Mn sublattice coexists with the ferromagnetic Tb sublattice below $T_C^{(Tb)} = 65$ K and a canted ferrimagnetic structure is stable below 53 K. The differences in the magnetic properties of the two compounds with X = Ge and Si can be attributed to different Mn–Mn distances. However, below $T_C^{(R)}$ the $f-d$ exchange interaction comes into play as well. The role of this factor is still unclear.

Considering the large difference between the lattice parameters of $TbMn_2Ge_2$ ($a=3.990$ Å, $c=10.845$ Å) and $TbMn_2Si_2$ ($a=3.930$ Å, $c=10.486$ Å)[1] a comparison of the magnetic characteristics of two systems $Tb_{1-x}Y_xMn_2Ge_2$ and $Tb_{1-x}Y_xMn_2Si_2$ with a variable $f-d$ exchange can be informative in clarifying the relative role of the factors, such as $f-d$ exchange interaction and interatomic distances, in the

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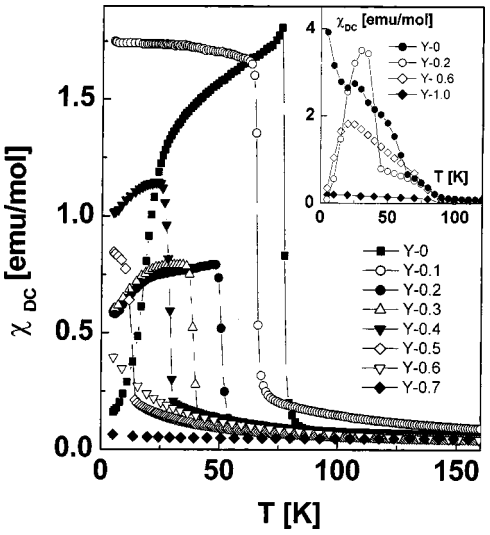

FIG. 1. Temperature dependence of the dc susceptibility of the $Tb_{1-x}Y_xMn_2Ge_2$ compounds measured at 1000 Oe. The inset shows the same data for the $Tb_{1-x}Y_xMn_2Si_2$ system.

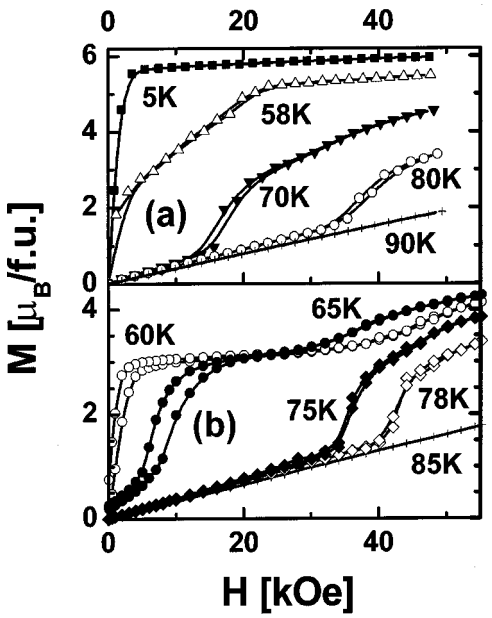

FIG. 2. Magnetization curves of $TbMn_2Si_2$ (a) and $Tb_{0.9}Y_{0.1}Mn_2Ge_2$ (b).

low temperature behavior of the RMn_2X_2 intermetallics.

In this work, the temperature- and field-induced magnetic phase transitions have been studied on textured samples of the pseudoternary $Tb_{1-x}Y_xMn_2X_2$ systems by magnetization and thermal expansion measurements.

EXPERIMENT

The polycrystalline samples have been synthesized by induction melting of elemental materials and homogenized for 170 h. at 750 °C. The magnetization in the temperature range 4–400 K has been measured by a superconducting quantum interference device (SQUID) magnetometer in fields up to 55 kOe. The thermal expansion has been studied in the temperature range 8–300 K by the x-ray diffraction method using a He flow cryostat.

In Fig. 1 the temperature dependence of the dc susceptibility χ_{DC}, is shown for the $Tb_{1-x}Y_xMn_2Ge_2$ compounds. The sharp increase of χ_{DC} is due to the onset of magnetic order in the RE sublattice. $T_C^{(Tb)}=76$ K for $x=0$ and decreases with increasing Y concentration. The low-temperature transition from a canted to a collinear ferromagnetic structure known for $TbMn_2Ge_2$ can be associated with the rapid change of χ_{DC} at 18 K. Similar dependencies, although more smoothed at $T_C^{(Tb)}$, have been obtained for the $Tb_{1-x}Y_xMn_2Si_2$ system (see inset in Fig. 1). Note, however, the shape of $\chi_{DC}(T)$ near the transition temperature depends strongly on texture in both systems (compare also with Refs. 4 and 10). More reliably the transition temperatures can be determined from the magnetization measurements.

As can be seen from Fig. 2(a), where the results for $TbMn_2Si_2$ are exemplified, the antiferromagnetic (70, 80, and 90 K), canted (58 K), and collinear (5 K) ferromagnetic phases can be distinguished by the character of magnetization process. In the vicinity of the phase transition the magnetic field induces transitions between the antiferromagnetic and canted phases and between the canted and collinear phases. Hence, the transition temperatures $T_C^{(Tb)}$ and T_{tr} can be evaluated extrapolating the critical fields of the transitions to zero. Similar transitions occur in the $Tb_{1-x}Y_xMn_2Ge_2$ system [Fig. 2(b)].

Using the magnetization data, the magnetic phase diagrams of the $Tb_{1-x}Y_xMn_2X_2$ systems have been constructed (Fig. 3). The two diagrams practically coincide with each other, the scattering of the transition points $T_C^{(Tb)}$ and T_{tr} between $TbMn_2Ge_2$ and $TbMn_2Si_2$ being within the range reported in the literature. In the Y-rich region, $x>0.5$, no transitions are observed in the magnetization curves. The temperature dependence of the low-field susceptibility varies linearly below about 170 K in all these compounds. This indicates that the Tb sublattice is in the paramagnetic state for $x>0.5$.

The temperature dependence of the unit cell volume of typical $Tb_{1-x}Y_xMn_2X_2$ compounds is given in Fig. 4. Only two samples, $TbMn_2Ge_2$ and $Tb_{0.9}Y_{0.1}Mn_2Ge_2$, show a pronounced volume expansion at $T_C^{(Tb)}$. The volume effect

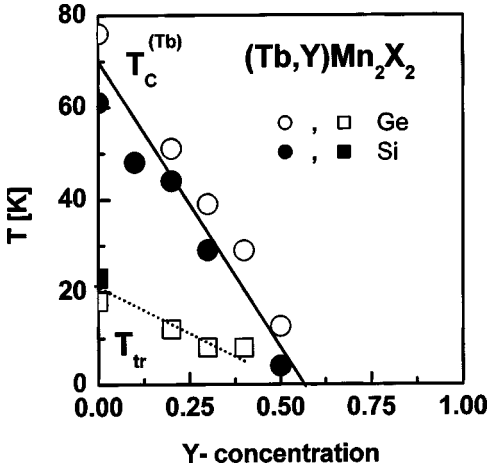

FIG. 3. Magnetic $x-T$ phase diagrams of the $Tb_{1-x}Y_xMn_2Ge_2$ and $Tb_{1-x}Y_xMn_2Si_2$ systems.

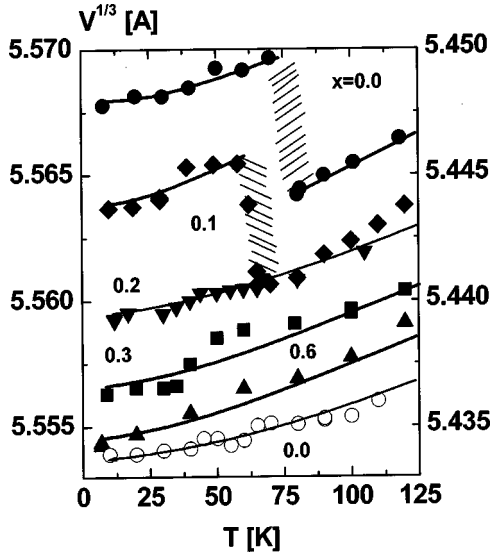

FIG. 4. Temperature dependence of the unit cell parameter $V^{1/3}=(a^2c)^{1/3}$ for some characteristic samples of $Tb_{1-x}Y_xMn_2Ge_2$ (full symbols, left scale) and $TbMn_2Si_2$ (open symbols, right scale). The hatched regions show the area of coexistence of the phases in two Ge-containing compounds with a volume expansion at $T_C^{(Tb)}$.

$\Delta V/V$ is equal to 3.2×10^{-3} and 2.7×10^{-3} for $x=0.0$ and 0.1, respectively. It is important to note that the transition at $T_C^{(Tb)}$ is of a first-order type according to the x-ray χ_{DC} and $M(H)$ data.

Due to the large volume effect, coexistence of the antiferromagnetic and canted ferromagnetic phases has been observed in the vicinity of $T_C^{(Tb)}$ in $TbMn_2Ge_2$ and $Tb_{0.9}Y_{0.1}Mn_2Ge_2$ by x-ray diffraction. The intensity of the x-ray peaks belonging to these phases varies smoothly with temperature according to the fraction of each phase. Such an overlapping of the diffraction peaks near $T_C^{(Tb)}$ must be observable in the neutron diffraction experiments, too.

CONCLUDING REMARKS

As can be seen from Fig. 3, the magnetic ordering of the Tb sublattice breaks at the same Y concentration for both X=Ge and Si. The fact that the lattice parameters are different in two systems indicates that the main factor in stabilizing a magnetic order in the Tb sublattice is the $f-d$ exchange interaction. One can suggest that the $f-d$ molecular field acting on Tb is weak in the RMn_2X_2 series and is canceled out above $T_C^{(Tb)}$ because of the negative Mn–Mn exchange. A first-order type magnetic phase transition at $T=T_C^{(Tb)}$ takes place in these compounds because a spontaneous moment arises in the Mn sublattice due to the $f-d$ exchange. Hence, the total molecular field acting on the Tb sublattice changes abruptly at $T_C^{(Tb)}$.

The large volume expansion observed at $T_C^{(Tb)}$ in the Tb-rich $Tb_{1-x}Y_xMn_2Ge_2$ compounds requires study more in detail. This effect can depend on the details of the Mn sublattice magnetic structure below $T_C^{(Tb)}$ and can arise when the angle between the Mn spin moments exceeds some borderline value.

ACKNOWLEDGMENTS

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