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Structure and magnetic properties of $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds

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Abstract. $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds with $x = 0, 0.05, 0.10, 0.15, 0.20$ and 0.25 have been studied in this paper. All of the compounds crystallize in the $HfFe_6Ge_6$-type structure (space group $P6/mmm$). The substitution of Co for Mn in $TbMn_6Sn_6$ results in a monotonic decrease in the lattice constants and the unit-cell volume. The ordering temperature decreases monotonically from 423 K at $x = 0$ to 267 K at $x = 0.25$. The reorientation of the easy-axis magnetization of $TbMn_6Sn_6$ disappeared on Co substitution at $x = 0.10$, while for Co content x exceeding 0.15 a compensation temperature was observed in the thermal magnetization curve. The reorientation temperature decreases with Co content increase, while the compensation temperature increases. The magnetization curves up to 70 kOe at 4.5 K are given and the saturation magnetization decreases from 3.4 μ_B/f.u. at $x = 0$ to 0.2 μ_B/f.u. at $x = 0.1$ and then increases to 2.8 μ_B/f.u. at $x = 0.25$.

1. Introduction

In the recent years, new ternary compounds RT_6X_6 (T = Fe or Mn; X = Ge or Sn) have attracted great attention [1–4]. For RMn_6Sn_6 (R = Gd, Tb, Dy or Ho) evidence of ferrimagnetism has been reported. Both the R sublattice and the Mn sublattice show ferromagnetism at high temperatures. The magnetic structure of $TbMn_6Sn_6$ at 300 K consists of a stacking of ferromagnetic (001) layers of rare earth and manganese with the coupling sequence Mn(+)R(−)Mn(+)Mn(+)R(−)Mn(+). The magnetic moments are $\mu_{Mn} = 2.0\ \mu_B$, and $\mu_{Tb} = 4.9\ \mu_B$, which deviate from the c-axis with a deviation angle of 15°. At 2 K the magnetic moments are along [001] with $\mu_{Mn} = 2.4\ \mu_B$ and $\mu_{Tb} = 8.6\ \mu_B$ [2]. A second apparent transition was found to arise at 330 K correlating with a change of the easy-axis magnetization direction [1]. The moments lie in the basal plane at high temperature and are along or near c below the transition temperature. The effect of Ti substitution for Mn on the structure and magnetic properties of $TbMn_6Sn_6$ has been investigated and reported [5]. In the present paper, the structure and magnetic properties of pseudo-ternary $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds have been investigated in detail.

2. Experimental details

All of the compounds, each with a total mass of about 10 g, were prepared by argon arc melting from starting materials of at least 99.9% purity. The alloys were melted at least five

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times to ensure homogeneity. After arc melting, the polycrystalline specimens were sealed into evacuated quartz tubes and annealed at 1073 K for ten days or more and then quenched into water. The crystal structure was analysed by x-ray powder diffraction using Cu Kα radiation. The thermal magnetic behaviours from 5 K to 300 K were measured from free powder using a superconducting quantum interference device (SQUID) magnetometer in the field of 400 Oe. The temperature dependencies of the magnetization between 180–480 K at the magnetic field of 400 Oe were measured using a vibrating-sample magnetometer (VSM) and the ordering temperatures were derived from them. The specific magnetizations were measured for free powder in a field up to 70 kOe using a SQUID at 4.5 K. Due to the slope of the magnetization curves in the field up to 70 kOe, the saturation magnetizations at 4.5 K were obtained by extrapolating B to zero.

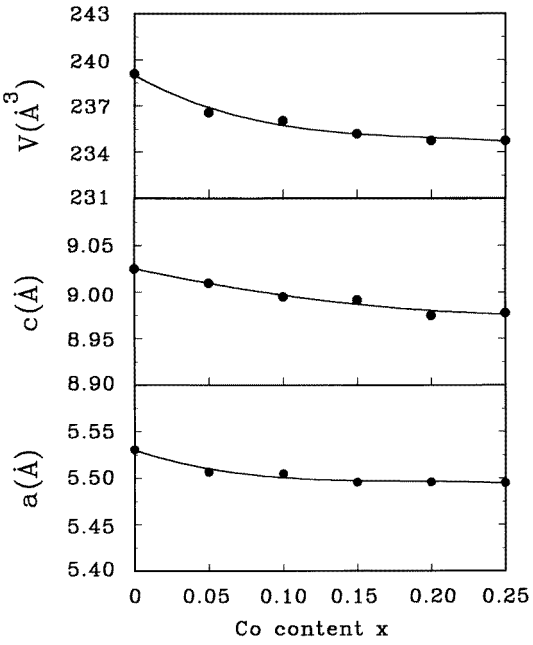

Figure 1. The dependence on Co content of the lattice constants a and c and the unit-cell volume V for the $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds.

3. Results and discussion

The crystal structures were analysed by x-ray powder diffraction using Cu Kα radiation. The results indicate that all of these compounds are single-phase materials, with $HfFe_6Ge_6$-type structure. The crystal structure of $HfFe_6Ge_6$ is a filled derivative of the CoSn B35-type structure (space group, $P6/mmm$). Neutron diffraction results have indicated that Tb atoms occupies the 1b sites, Mn atoms occupy the 6i sites, and Sn atoms occupy the 2e, 2d, and 2c sites [2]. Upon the substitution of Co for Mn, the structure is not changed; therefore, Co atoms also occupy 6i sites like Mn. The values of the lattice constants a and c and the unit-cell volume V are listed in table 1. The lattice constants a and c and unit-cell volume V as functions of the Co concentration are also shown in figure 1. It can be seen that the substitution of Co for Mn results in a monotonic decrease in the lattice constants and the

Table 1. The values of lattice constants a and c, the unit-cell volume V, the ordering temperature, the reorientation temperature, the compensation temperature, the saturation magnetization, and the magnetization of the transition metal sublattice at 4.5 K for the $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds.

x	a (Å)	c (Å)	V (Å^3)	T_C (K)	T_t (K)	T_k (K)	M_S (μ_B/f.u.)	M_T (μ_B/f.u.)
0.0	5.5306	9.0253	239.10	423	330		3.4	12.4
0.05	5.5065	9.0098	236.59	352	289		1.9	10.9
0.10	5.5050	8.9950	236.07	334			0.2	9.2
0.15	5.4961	8.9915	235.22	310		130	0.6	8.4
0.20	5.4959	8.9749	234.77	296		135	1.1	7.9
0.25	5.4951	8.9779	234.78	267		140	2.8	6.2

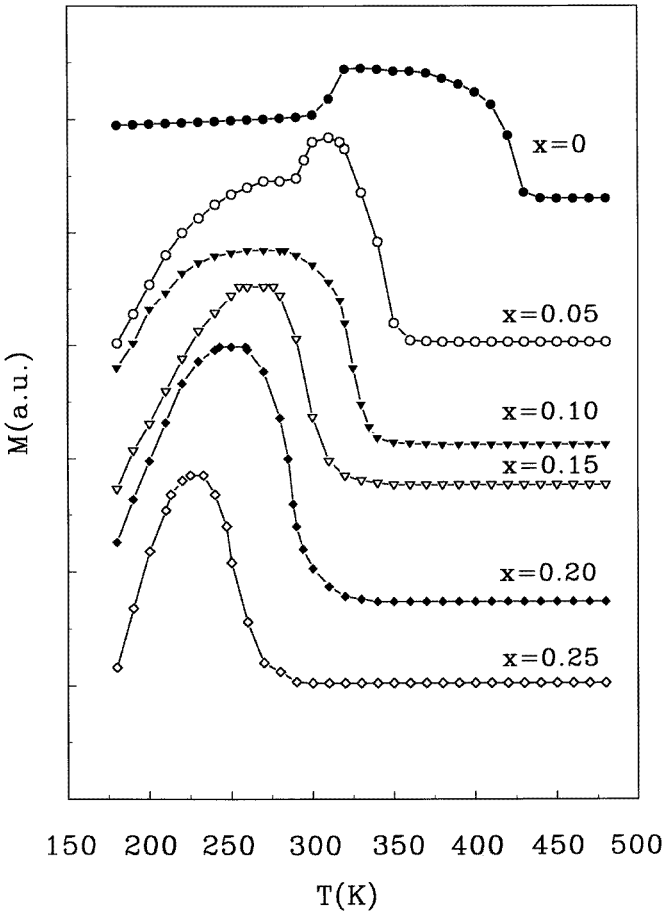

Figure 2. The thermal magnetic behaviour under a magnetic field of 400 Oe in the temperature range 180–480 K for $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds.

unit-cell volume with increasing Co concentration. This may be associated with the smaller radius of the Co atoms compared with that of the Mn atoms.

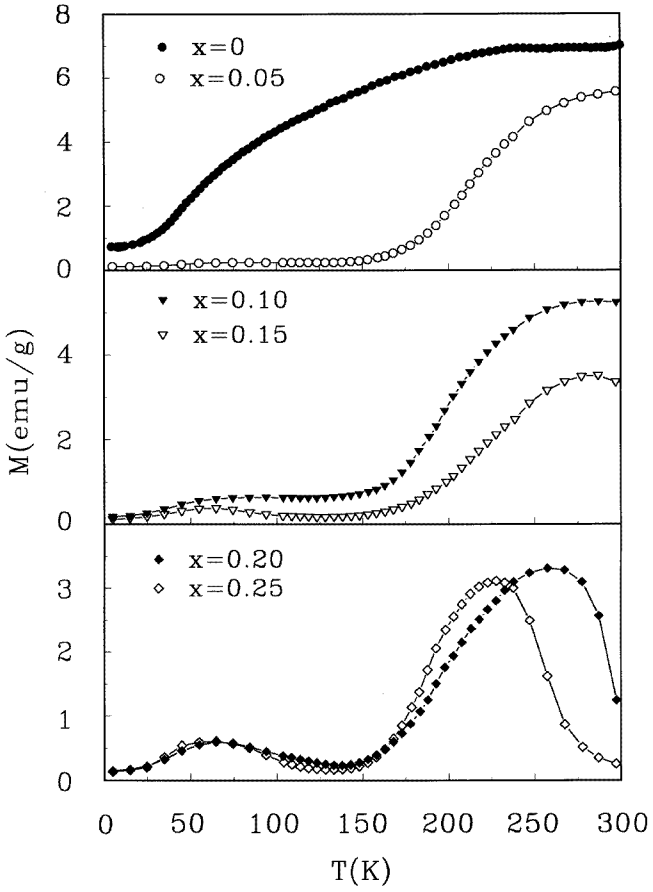

Figure 3. The thermal magnetic behaviour under a magnetic field of 400 Oe in the temperature range 5–300 K for $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds.

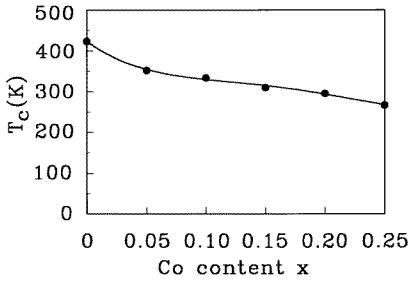

Figure 4. The ordering temperature changes as a function of Co concentration for the $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds.

The temperature dependencies of the magnetization of $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds are shown in figures 2 and 3. From the curves in figure 2 it can be found that the temperature of the transition correlated to the change of the easy-axis magnetization direction decreases

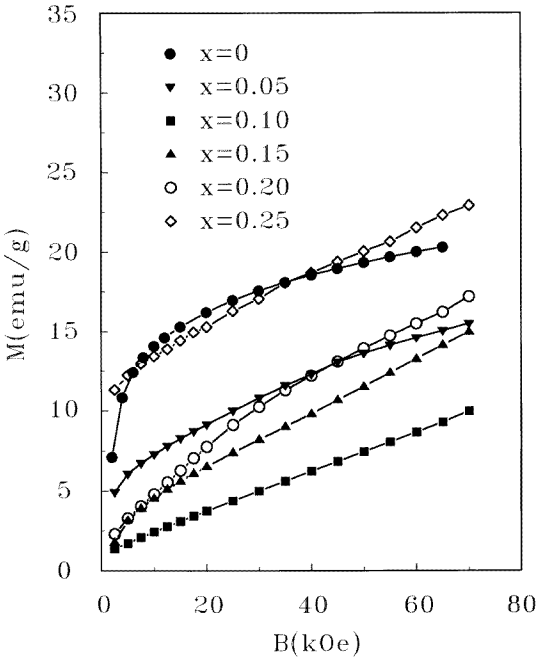

Figure 5. Magnetization curves for $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds at 4.5 K.

from 330 K at $x = 0$ to 289 K at $x = 0.05$, and that the transition disappeared when the Co content exceeded 0.1. Although it is not clear why the transition disappeared, this may be related to the decrease of the distance of the nearest Mn atoms and hence the enhancement of the exchange interaction of two Mn sublattices on Co substitution (T atoms—here Mn atoms—were considered as constituting two sublattices in the three-sublattice model [6, 7]). The ordering temperatures of these compounds can also be derived from these curves. A compensation temperature was found in the thermal magnetic curves in figure 3 for compounds with Co content x exceeding 0.15. This was due to the decrease of the Mn-sublattice moment on Co substitution as found for other series of R–T compounds [8, 9]. At higher Co concentration, the Mn-sublattice moment is lower than that of the Tb sublattice at 4.5 K. But, because the Tb-sublattice moment decreases more quickly as a function of temperature than the Mn-sublattice moment, they will become equal at a certain temperature. Data on the ordering temperature, the reorientation temperature and the compensation temperature are given in table 1. The ordering temperature changing as a function of Co concentration is shown in figure 4. It decreases monotonically from 423 K at $x = 0$ to 267 K at $x = 0.25$. This may be caused by the decrease of the magnetic moments of the Mn sublattice (see below).

The magnetization curves are shown in figure 5. According to the classification of Colpa and Brabers [7] the magnetization here seems to corresponding to a quasi-two-sublattice configuration for $x = 0$, changing to a three-sublattice configuration of type III-upper for $x = 0.25$. A value for the saturation magnetization can be obtained by extrapolation of the magnetization to $B = 0$. The values for saturation magnetization are listed in table 1. The saturation magnetization decreases from 3.4 μ_B/f.u. at $x = 0$ to 0.2 μ_B/f.u. at $x = 0.1$ and then increases to 2.8 μ_B/f.u. at $x = 0.25$ as shown in figure 6. The magnetic moment of the

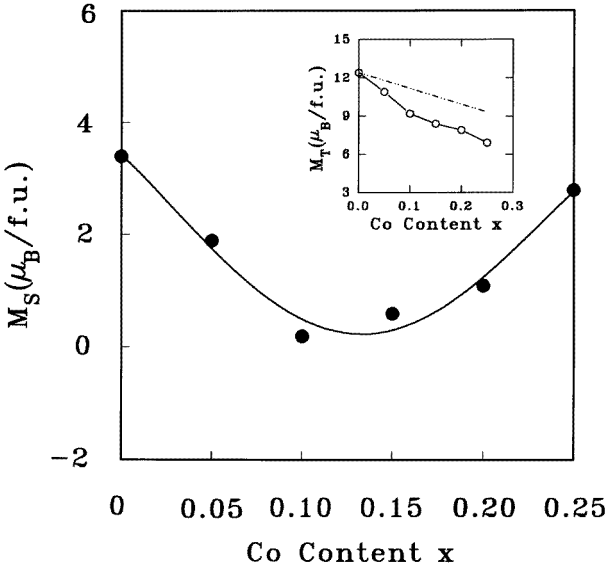

Figure 6. The Co content dependence of the saturation magnetization for $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds at 4.5 K; The inset shows the Co content dependence of the T-sublattice magnetization.

T sublattice (T stands for transition metal) is larger than that of the Tb sublattice for $x = 0$ and as x increases the opposite situation arises, so a minimum appears. An estimate can be obtained for the T-sublattice moment assuming the Tb moments to be equal to the free-ion moments. The estimated T-sublattice moment decreases as the Co concentration x increases. The inset in figure 6 compares the T-sublattice moment with the value of that expected for the case of simple magnetic dilution (the dashed-dotted curve). On Co substitution the exchange interaction is enhanced—as the nearest Mn–Mn distance decreases, so it seems impossible for the Mn moments to decrease. The fact that the concentration dependence falls below the simple dilution curve means that it has to be taken into account that the Co atoms carry magnetic moments which couple in an antiparallel fashion to the Mn moments in $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds. From the value for the pure $TbMn_6Sn_6$ a moment of 2.07 μ_B per Mn atom can be deduced. This means that, if in $Tb(Mn_{1-x}Co_x)_6Sn_6$ the Mn moment does not change when Co is substituted for Mn, and if the Co moments are coupled in an antiparallel fashion to the Mn moments, the average apparent magnetic moment of Co atoms has a value of about 2.5 μ_B.

4. Conclusion

In the present paper, a study of the structural and magnetic properties of $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds (x = 0–0.25) are reported. The main results are gathered together in table 1. The $Tb(Mn_{1-x}Co_x)_6Sn_6$ compounds crystallize in the $HfFe_6Ge_6$-type structure. The substitution of Co for Mn in $TbMn_6Sn_6$ decreases the lattice constants and the unit-cell volume from $a = 5.5306$ Å, $c = 9.0253$ Å, $V = 239.10$ Å^3 at $x = 0$ to $a = 5.4951$ Å, $c = 8.9779$ Å, $V = 234.78$ Å^3 at $x = 0.25$. The ordering temperature decreases monotonically from 423 K at $x = 0$ to 267 K at $x = 0.25$. The reorientation of

the easy-axis magnetization of $TbMn_6Sn_6$ disappeared on Co substitution with x exceeding 0.10, While for x exceeding 0.15 a compensation temperature was observed in the thermal magnetization curves. The reorientation temperature decreases with Co content increase, while the compensation temperature increases. The magnetization curves up to 70 kOe at 4.5 K are given and the saturation magnetizations are derived from them. The saturation magnetization decreases from 3.4 μ_B/f.u. at $x = 0$ to 0.2 μ_B/f.u. at $x = 0.1$ and then increases to 2.8 μ_B/f.u. at $x = 0.25$. The decrease of the T-sublattice moment was explained by assuming that Co has a magnetic moment of about 2.5 μ_B per atom and is coupled in an antiparallel fashion with Mn.

Acknowledgments

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