Structure and magnetic properties of $TbMn_{6-x}V_xSn_6$ compounds

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Abstract. In the present work, the influence of substitution of V for Mn on the structure and magnetic properties of $TbMn_{6-x}V_xSn_6$ compounds has been investigated. The lattice constants, the Curie and spin-reorientation temperatures, the anisotropy field and the saturation magnetization have been determined as a function of the V content.

1. Introduction

Recently, new ternary compounds RT_6X_6 (T = Fe or Mn, X = Sn or Ge) have attracted much attention [1–8]. RMn_6Sn_6 (R = Gd, Dy, Tb or Ho) are found to be ferrimagnetic [2, 3], while both the R and the Mn sublattices show ferromagnetic behaviour at high temperatures. A large coercivity of 1.1 T has been found in the compound $TbMn_6Sn_6$ at 4.2 K [2]. With increasing temperature from 77 to 300 K, the coercivity first increases, going through a maximum with coercivity 1.2–1.3 T at about 200 K, and then decreases [7]. The magnetic structure of $TbMn_6Sn_6$ at 300 K consists of a stacking of ferromagnetic (001) layers of rare earth and manganese with coupling sequence Mn(+)R(−)Mn(+)Mn(+)R(−)Mn(+). At 2 K the magnetic moments are along the [001] direction [3]. A phenomenon of spin reorientation was found at about 330 K [2, 7]. The magnetic moments lie in the basal plane at high temperatures and along or near the c-axis below the spin-reorientation temperature. Element substitution is usually carried out not only to improve the performance of magnetic materials, but also to examine the fundamental magnetic properties. In the present paper, the influence of the substitution of V for Mn on the structure and the magnetic properties of $TbMn_6Sn_6$ has been investigated in detail.

2. Experimental details

$TbMn_{6-x}V_xSn_6$ compounds with $x = 0$, 0.35, 0.5, 1.0, 1.5 and 2.0 were prepared by arc-melting the starting elements of at least 99.9% purity in an Ar atmosphere. The ingots were remelted several times to ensure homogeneity. After arc-melting, the polycrystalline specimens were sealed into evacuated quartz tubes and annealed at 1073 K for 2 weeks

or more and then quenched into water. The crystal structure was analysed by means of x-ray diffraction using Cu Kα radiation. The thermal magnetic behaviour was measured in a magnetic field of 0.05 T in the temperature range from 100 to 450 K, using a vibrating sample magnetometer (VSM). In order to obtain the aligned samples, fine powders of the sample were mixed with epoxy resin and bonded in a magnetic field of 1 T for one day. The magnetic anisotropy fields were measured with a high-pulsed-magnetic-field installation at room temperature in external fields applied in the direction perpendicular to the easy axis of the TbMn$_{6-x}$V$_x$Sn$_6$ aligned samples by the singular-point detection (SPD) technique [9]. The magnetization curves of powders were also measured and values of saturation magnetization at room temperature were determined from M–$1/B$ plots by extrapolating $1/B$ to zero.

3. Results and discussion

Results of x-ray diffraction patterns of powder samples show that all the TbMn$_{6-x}$V$_x$Sn$_6$ compounds investigated crystallize in the HfFe$_6$Ge$_6$-type structure. The crystal structure of HfFe$_6$Ge$_6$ is a filled derivative of the CoSn B35-type structure (space group, $P6/mmm$) [1, 3]. All these samples are almost single phase. The lattice constants a, c and the unit-cell volume v derived from the x-ray diffraction patterns are listed in table 1. It can be seen that with increasing V content in compounds up to $x = 0.5$, the lattice constant a varies very little and the lattice constant c increases. With further increase of V content in the range $x = 0.5$–2.0, the constant a increases, while the constant c decreases first and finally increases slightly. The unit-cell volume v increases monotonically with increasing V content from $x = 0$ to 2.0.

Table 1. Lattice constants a, c, unit-cell volume v, Curie temperature T_c, spin-reorientation temperature T_{sr}, magnetic anisotropy field B_a and saturation magnetization M_s at room temperature for TbMn$_{6-x}$V$_x$Sn$_6$ compounds.

x	a (Å)	c (Å)	v (Å^3)	T_c (K)	T_{sr} (K)	B_a (T)	M_s (A m^2 kg^{-1})
0.0	5.5276	9.0282	238.9	421	320	0.80	22.54
0.35	5.5258	9.0395	239.0	413	317	0.75	19.05
0.5	5.5271	9.0434	239.3	400	313	0.72	18.16
1.0	5.5311	9.0358	239.4	410	308	0.37	17.59
1.5	5.5338	9.0323	239.5	420	306	0.34	15.06
2.0	5.5383	9.0338	240.0	420	300	0.28	12.55

The results of the thermomagnetic behaviour of the TbMn$_{6-x}$V$_x$Sn$_6$ compounds are shown in figure 1. All compounds have the same behaviours as the $x = 0$ one, where the ferrimagnetic structure has been established [3]. Figure 2 shows the V content dependences of the Curie temperature and lattice constant c for TbMn$_{6-x}$V$_x$Sn$_6$ compounds. As the V content increases, the Curie temperature T_c of TbMn$_{6-x}$V$_x$Sn$_6$ compounds decreases very fast in the composition range $x = 0$–0.5, then increases again in the range $x = 0.5$–1.5, and finally remains the same value in the range $x = 1.5$–2.0. It seems that the V content dependence of Curie temperature may be associated with the V content dependence of crystal constant c in the TbMn$_{6-x}$V$_x$Sn$_6$ compounds.

It can be seen from the thermomagnetic curves shown in figure 1 that there is an abrupt increase of the magnetization in the thermomagnetic curves at certain temperatures T_{sr},
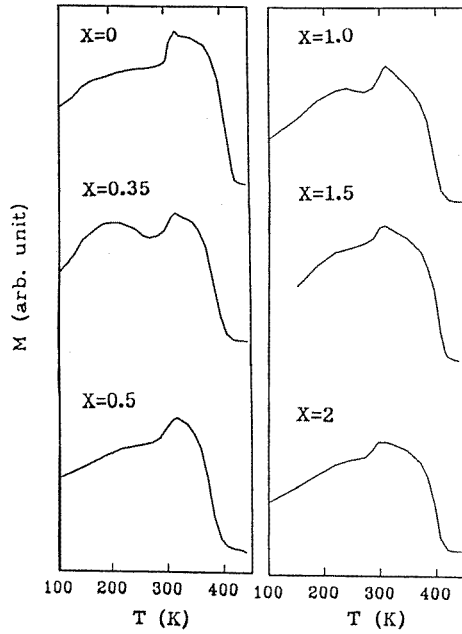
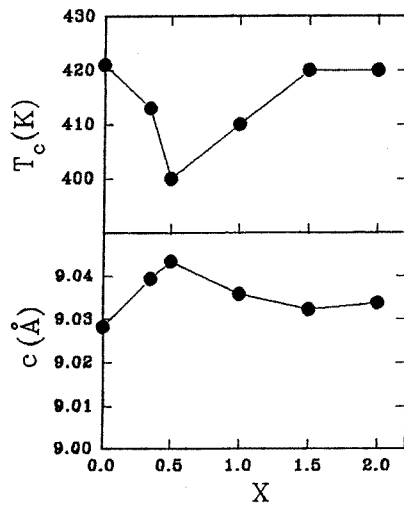**Figure 1.** Thermomagnetic curves in a field of 0.05 T for TbMn$_{6-x}$V$_x$Sn$_6$ compounds.

Figure 2. The V content dependences of the Curie temperature T_c and the lattice constant c for TbMn$_{6-x}$V$_x$Sn$_6$ compounds.

which depend on the V content. Such an abrupt increase of the magnetization indicates occurrence of the spin reorientation [2, 3, 7]. Neutron diffraction analysis of TbMn$_6$Sn$_6$ [3] showed that there is a spin reorientation at about 300 K. At temperature lower than 300 K, a collinear ferrimagnetic arrangement along the *c*-axis exist for the moments of Tb and Mn due to the antiferromagnetic coupling between Tb and Mn sublattices. At temperature

above 300 K moments of Tb and Mn lie in the basal plane. It can be seen from table 1 that the spin-reorientation temperature T_{sr} decreases monotonically with increase of the V content for the $TbMn_{6-x}V_xSn_6$ compounds.

The values of the magnetocrystalline anisotropy field B_a for the $TbMn_{6-x}V_xSn_6$ compounds are listed in table 1. It can be seen that with increasing V content the anisotropy field decreases monotonically. In a previous work [8] it has been found that the spin-reorientation temperature and the magnetic anisotropy field of $TbMn_{6-x}Al_xSn_6$ compounds first decrease with increasing Al content, go through a minimum at $x = 0.5$, then increase with further increasing Al content. In the present work the anisotropy field B_a and spin reorientation T_{sr} are also found to have similar dependence on the V content in $TbMn_{6-x}V_xSn_6$ compounds. In the $TbMn_{6-x}V_xSn_6$ compounds, the magnetic anisotropy results from the contribution of Tb and Mn sublattices. The total anisotropy constant K may be simply expressed as $K = K_{Tb} + K_{Mn}$ and the spin reorientation results from the competition between K_{Tb} and K_{Mn}. At temperature below T_{sr} the magnetic moments lie along the c-axis and the total magnetic anisotropy is dominated by the term K_{Tb} which prefers easy-axis anisotropy. At temperature above T_{sr}, the moments lie in the basal plane and the total magnetic anisotropy is dominated by the term K_{Mn} which prefers planar anisotropy. The decrease of B_a with increasing V content implies that K_{Tb} decreases with V content which leads to a decrease of spin-reorientation temperature T_{sr}.

The values of the saturation magnetization M_s for the $TbMn_{6-x}V_xSn_6$ compounds are also listed in table 1. It can be seen that with increasing V content, the saturation magnetization decreases monotonically, which may be associated with the V atoms being non-magnetic.

Acknowledgment

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