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Magnetic properties of $R_2Co_{17-x}Ga_x$ and $RCo_{5-x}Ga_x$ alloys (R = Y, Tb) in high magnetic fields

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Magnetic properties of $R_2Co_{17-x}Ga_x$ and $RCo_{5-x}Ga_x$ alloys, where R = Y and Tb, have been investigated in steady and pulsed magnetic fields up to 14 and 34 T, respectively. For the $Y_2Co_{17-x}Ga_x$ alloys saturation is reached in relatively low magnetic fields and the saturation magnetic moment decreases with increasing Ga content. No saturation was observed for the $YCo_{5-x}Ga_x$ alloys and the Curie temperatures were lower than for the corresponding 2:17 phases. The Curie temperatures of the $TbCo_{5-x}Ga_x$ samples were also lower than for the corresponding 2:17 materials. The Tb compounds did not reach saturation even in the highest magnetic field. The minimum in the temperature dependence of the magnetization is an indication of ferrimagnetic character. The substitution of Ga for Co in $TbCo_{5-x}Ga_x$ increases the magnetic moment, which may be explained as a result of the replacement of Co atoms with magnetic moments antiparallel to those of the Tb atoms.

Rare-earth alloys and compounds / Magnetic measurements / Magnetization

Introduction

During the past decades the structure and intrinsic magnetic properties of R_2T_{17} (R = rare earth, T = Fe, Co) compounds have been extensively studied because of the promising magnetic properties of these materials and their potential application as materials for hydrogen storage, etc. A strong uniaxial magnetic anisotropy is required to achieve high coercivity, but the binary R_2Co_{17} compounds exhibit uniaxial anisotropy only for R = Sm, Er and Tm. There exists a possibility that substitution of Ga for Co can induce uniaxial anisotropy in the compounds formed by other rare earths. The structure and magnetic properties of the Tb and Y compounds have recently been examined [1–4]. The crystal structure of these extended solid solutions depends on the Ga concentration. A preliminary examination of the Tb compounds revealed only the existence of a rhombohedral Th_2Zn_{17}-type phase [1,2], however, a more detailed investigation of this system showed that with increasing Ga concentration the following sequence of phases exists: Th_2Ni_{17} (hex) → Th_2Zn_{17} (rhomb) [4]. The Y system exhibits a still more complicated situation: Th_2Ni_{17} (hex) → $TbCu_7$ (hex) → Th_2Zn_{17} (rhomb) [3]. In both $Tb_2Co_{17-x}Ga_x$ [1,2] and $Y_2Co_{17-x}Ga_x$ [3] an increase of the Ga concentration causes a decrease of the Curie temperature, whereas the same effect on the saturation magnetization was observed only in the Y system. In both systems saturation was reached in relatively low magnetic fields.

In the present study, we have extended the investigation of the magnetic properties to measurements of the magnetization at low temperature in high (up to 34 T) pulsed magnetic fields, with the aim of checking if high magnetic fields can influence the mutual coupling of the Tb and Co magnetic sublattices. The results obtained for the Y alloys should serve as a reference for the magnetic Co sublattice. Magnetic measurements for the $RCo_{5-x}Ga_x$ phases that were discovered in the Tb-Co-Ga and Y-Co-Ga systems are also reported.

Experimental details

$R_2Co_{17-x}Ga_x$ samples of nominal composition $Y_2Co_{17-x}Ga_x$, x = 0, 0.9, 1.9, 3.23, 4.75, 5.7, 8.55 [3] and $Tb_2Co_{17-x}Ga_x$, x = 0, 1.9, 2.85, 3.8, 8.55 [4] were investigated. For the investigation of the $RCo_{5-x}Ga_x$ phases with the closely related hexagonal structure type $CaCu_5$, the following compositions were selected: x = 0.6 and 1.5 for $YCo_{5-x}Ga_x$, and x = 0.6, 0.9 and 1.2 for $TbCo_{5-x}Ga_x$. All samples were prepared by arc-melting the constituents under high-purity argon (99.998 %) on a water-cooled cooper hearth. The ingots were then heat treated in evacuated silica tubes at 870 K for two weeks and subsequently quenched in cold water (still in the silica tubes). Phase analysis was carried out using X-ray diffraction and the samples were further characterized by Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray analysis (EDX). All the samples were found to be single-phase materials. The magnetic measurements were carried out on polycrystalline pieces.

The field dependence of the magnetization at 4.2 K in steady magnetic fields was measured up to 14 T using a string magnetometer (not shown) and up to 35 T in pulsed magnetic fields (pulse duration 10 ms) [5]. The temperature dependence of the magnetization at T = 1.9-400 K was measured in a magnetic field of 0.5 T with a SQUID magnetometer, and in a magnetic field of 0.483 T at increasing temperature up to 1100 K with a home made installation.

Results and discussion

Crystallographic data for the $R_2Co_{17-x}Ga_x$ samples and results of magnetic measurements in low magnetic fields were published in [3,4]. Cell parameters of the $RCo_{5-x}Ga_x$ samples are listed in Table 1 and Table 2. The magnetic field dependence of the magnetization (magnetic moment) at 4.2 K for the $Y_2Co_{17-x}Ga_x$ and $YCo_{5-x}Ga_x$ alloys in magnetic fields up to 35 T is shown in Fig. 1. It can be seen that for the $Y_2Co_{17-x}Ga_x$ alloys (solid lines) saturation is reached in relatively low magnetic fields. In high magnetic fields the magnetic moment of the pristine Y_2Co_{17} compound is lower than the moments of the alloys with x = 0.9 and x = 3.23. We have so far found no explanation for this behavior. The magnetization *vs.* magnetic field curves obtained for the $YCo_{5-x}Ga_x$ alloys (dashed lines in Fig. 1) show that saturation is more difficult to achieve than for the $Y_2Co_{17-x}Ga_x$ alloys. No saturation was observed for the compound

Table 1 Lattice parameters and magnetic data for $TbCo_{5-x}Ga_x$ alloys with $CaCu_5$-type structure.

x	Lattice parameters		μ_s [a]	T_{max}
	a (Å)	*c* (Å)	(μ_B/f.u.)	(K)
0.6	4.969(2)	4.013(2)	5.0	530
0.9	4.9721(8)	4.0286(8)	5.4	500
1.2	4.984(2)	4.036(3)	6.2	400

[a] "saturation" magnetic moment at T = 4.2 K in magnetic field $\mu_0 H$ = 34 T

Table 2 Lattice parameters and magnetic data for $YCo_{5-x}Ga_x$ alloys with $CaCu_5$-type structure.

x	Lattice parameters		μ_s [a]	T_{max}	T_C
	a (Å)	*c* (Å)	(μ_B/f.u.)	(K)	(K)
0.6	4.9787(6)	3.9894(9)	7.78	400	738
1.5	4.898(3)	4.120(3)	3.59		31120

[a] "saturation" magnetic moment at T = 4.2 K in magnetic field $\mu_0 H$ = 34 T

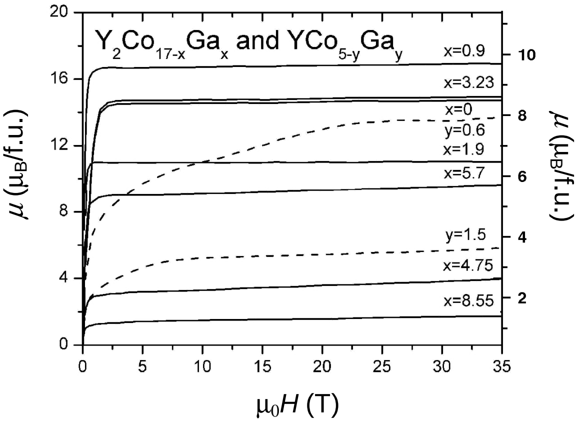

Fig. 1 Magnetic moment of $Y_2Co_{17-x}Ga_x$ and $YCo_{5-y}Ga_y$ *vs.* pulsed magnetic field at 4.2 K. The solid lines are for the former alloys and the dashed lines for the latter ones.

with x = 0.6, whereas for x = 1.5 saturation was reached only in a relatively high magnetic field. The sample with the higher Ga content has a lower value of magnetization.

Fig. 2 presents details of the magnetization *vs.* magnetic field curves for the $YCo_{5-x}Ga_x$ solid solution below 5 T at 1.9 and 4.2 K. Neither hysteresis nor remanence were observed and the two curves present similar features. However, the two alloys with different Ga concentration exhibit slightly different temperature dependences of the magnetic moment, as can be seen in Fig. 3, which shows the temperature dependence in a magnetic field of 0.5 T. For the material with higher Ga concentration the magnetization decreases with increasing temperature and the Curie point is at about 300 K, whereas for the sample with x = 0.6 the Curie temperature is about 750 K and a maximum at about 400 K is observed for the magnetization curve.

Fig. 4 shows the temperature dependence of the magnetic moment for the $TbCo_{5-x}Ga_x$ alloys. The magnetic moment decreases with increasing Ga concentration and the same is true for the temperature of the magnetic moment maximum. This type of behavior with a compensation point indicates the presence of two magnetic sublattices with oppositely directed magnetic moment (ferrimagnetic order).

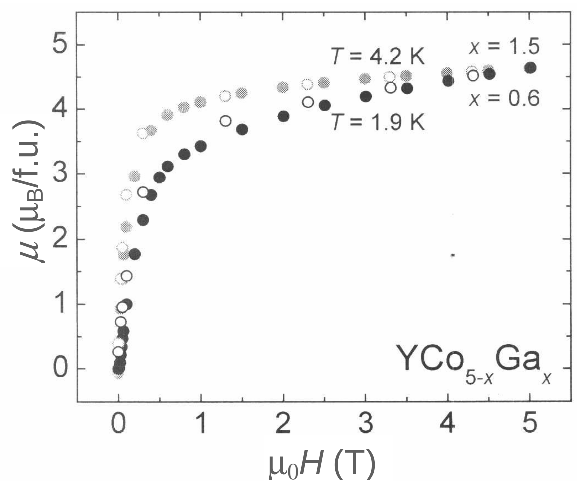

Fig. 2 Magnetic moment of $YCo_{5-x}Ga_x$ *vs.* steady magnetic field at 1.9 K (black filled circles: x = 0.6, black empty circles: x = 1.5) and 4.2 K (gray filled circles: x = 0.6, gray empty circles: x = 1.5).

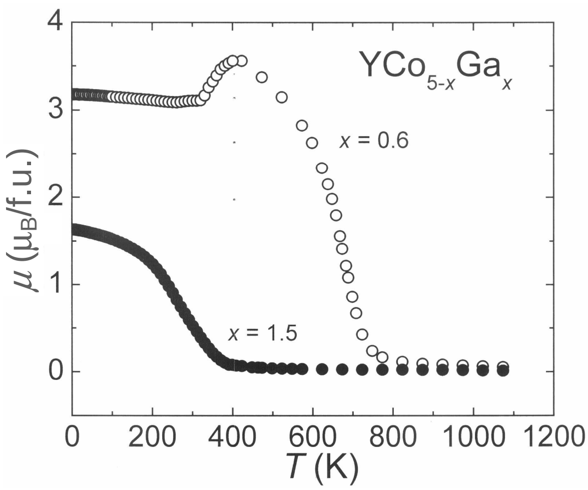

Fig. 3 Magnetic moment of $YCo_{5-x}Ga_x$ *vs.* temperature in a magnetic field of 0.5 T.

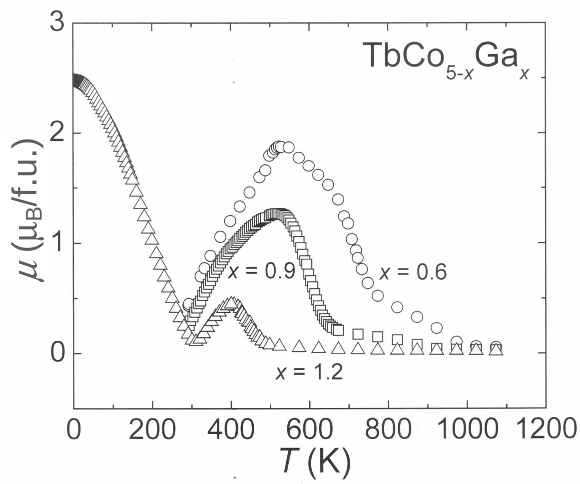

Fig. 4 Magnetic moment of $TbCo_{5-x}Ga_x$ *vs.* temperature in a magnetic field of 0.5 T.

Fig. 5 shows the magnetization *vs.* the magnetic field for $\mu_0 H \leq 5$ T for the solid solution $TbCo_{5-x}Ga_x$ at 1.9 K. One can see that in this magnetic field saturation is not reached. Moreover, all three alloys exhibit a small hysteresis, which is most pronounced for the sample with the smallest Ga content (x = 0.6). The magnetization is lowest for the same sample, whereas at higher temperature this sample shows the highest magnetization (see Fig. 4). The clear indication of opposite directions of the magnetization in the Tb and Co sublattices motivated us to apply higher magnetic fields. The Y phases were measured as reference system. Fig. 6 shows magnetization *vs.* the pulsed magnetic field for the $Tb_2Co_{17-x}Ga_x$ and $TbCo_{5-x}Ga_x$ alloys. In no case saturation was observed. The saturated magnetic moment computed per Tb atom for roughly the same Ga concentration (x = 0.9 for the 1:5 and x/2 = 0.95 for the 2:17 phase) is higher for the 1:5 phase (5.4 and 4.5 μ_B/Tb atom, respectively). Another interesting observation is the sequence of the values of the highest ("saturation") magnetic moment with respect to the Ga content. For the 2:17 alloys the "saturation" value of the magnetic moment decreases with increasing Ga concentration. This behavior may suggest that the Ga atoms substitute for Co atoms that do not contribute to the magnetic order antiparallel to the Tb sublattice. On the contrary, substitution of Ga for Co in the 1:5 intermetallics increases the total magnetic moment, which may result from the selective replacement of Co atoms with magnetic moments antiparallel to those of the Tb sublattice.

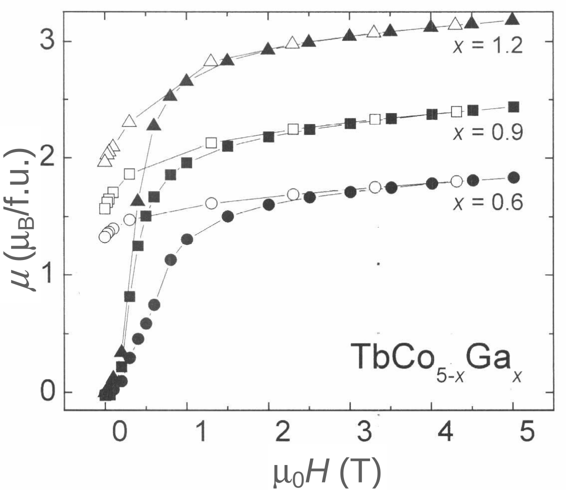

Fig. 5 Magnetic moment of $TbCo_{5-x}Ga_x$ *vs.* steady magnetic field at 1.9 K (filled symbols: increasing field, empty symbols: decreasing field).

Conclusions

The magnetic properties of the investigated intermetallics are complex. $Y_2Co_{17-x}Ga_x$ alloys can be saturated in relatively low magnetic fields and the saturation magnetic moment decreases with increasing

Ga concentration. For the $YCo_{5-x}Ga_x$ samples no saturation was detected and their Curie points are lower than for the 2:17 phases. For the $TbCo_{5-x}Ga_x$ samples the Curie points are also lower than for the corresponding 2:17 phases. The minimum observed in the temperature dependence of the magnetization for the Tb materials is an indication of ferrimagnetism.

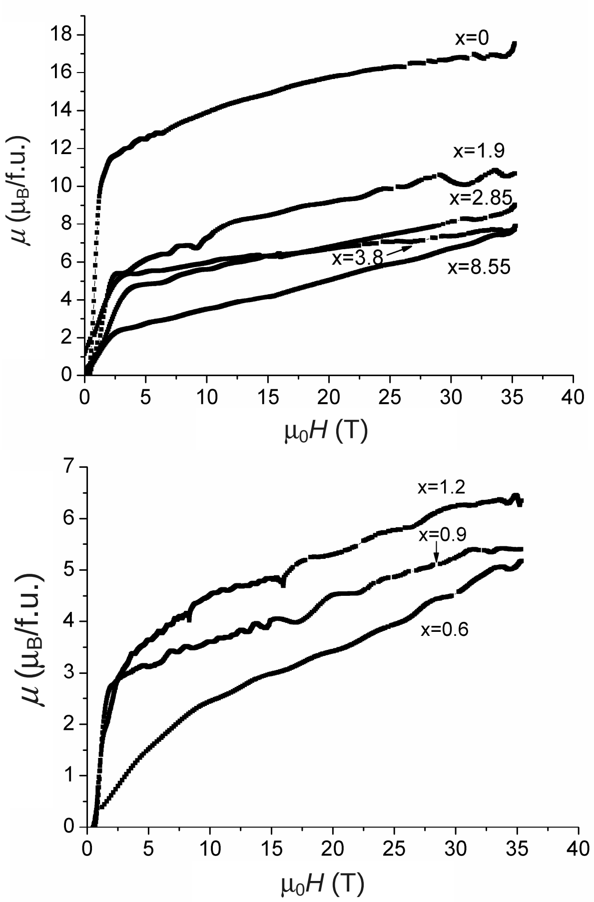

Fig. 6 Magnetic moment of $Tb_2Co_{17-x}Ga_x$ (a) and $TbCo_{5-x}Ga_x$ (b) *vs.* pulsed magnetic field at 4.2 K.

None of the Tb samples reached saturation in the highest applied magnetic field (35 T). In the $TbCo_{5-x}Ga_x$ phases the substitution of Ga for Co increases the magnetic moments with respect to the pristine compound. This is most probably the result of a selective replacement of Co atoms with magnetic moments antiparallel to those of the Tb atoms.

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