Journal of Magnetism and Magnetic Materials 177–181 (1998) 1044–1045

ELSEVIER

Journal of magnetism and magnetic materials

# Neutron diffraction and magnetization investigation of $Tb_2Co_{17-x}Ga_x$ intermetallics

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**Abstract**

The crystallographic and magnetic structures of $Tb_2Co_{17-x}Ga_x$ ($x = 1$, 3, 5 and 7) intermetallics have been investigated by time-of-flight neutron powder diffraction at 293 and 4.2 K. All four compounds crystallize in the rhombohedral $Th_2Zn_{17}$ structure (R $\bar{3}$ m) with Co atoms fully occupying the 9d site whilst Co and Ga atoms are found to distribute themselves in a non-random manner on the remaining 6c, 18f and 18h sites. These observed site occupations can be correlated with the sign reversal of the Co sublattice magneto-crystalline anisotropy for Ga concentrations $x > 3$. The magnetic structures of the Tb and Co sublattices have also been refined from the neutron-diffraction data and indicate a complex ferrimagnetic behaviour with Ga composition. The Co moment decreases in size with Ga concentration, as does the Curie temperature. 

*Keywords:* Neutron diffraction; Rare-earth intermetallics; Magnetic structure; Magnetocrystalline anisotropy

Intermetallic compounds which crystallize in the rhombohedral $Th_2Zn_{17}$ structure (space group R $\bar{3}$ m) [1] are found to display a large variation in magnetic properties such as magnetic-ordering temperatures and magneto-crystalline anisotropy. Recent investigations of the magnetic properties of $R_2Co_{17-x}Ga_x$ (R = Gd, Sm) compounds have demonstrated that Ga substitution can lead to a sign reversal of the magneto-crystalline Co sublattice anisotropy from easy plane to easy axis [2]. In contrast, $Dy_2Co_{17-x}Ga_x$ compounds are reported to have an easy-plane anisotropy across the whole concentration range [3]. In the present investigation, magnetic measurements have been extended to the $Tb_2Co_{17-x}Ga_x$ system whilst, via neutron diffraction, the underlying Co/Ga site occupation of the four transition-metal sublattices has been accurately quantified. A knowledge of these site occupancies is important for establishing any possible relation between the preferential site distribution of Ga atoms and the observed sign reversal of the Co sublattice anisotropy with Ga concentration.

Neutron-diffraction experiments were performed at room temperature and 4.2 K on the time-of-flight neutron-powder diffractometer ROTAX installed at the ISIS pulsed neutron source, Didcot, UK [4]. Data were collected simultaneously from multidetectors positioned in both backscattering ($2\theta = 124.5°$) and forward scattering ($2\theta = 37.5°$) banks. Samples of $Tb_2Co_{17-x}Ga_x$ were prepared by arc melting starting materials of Tb, Co and Ga with at least 99% purity. They were subsequently annealed in an evacuated quartz tube at 900 K for several weeks. All the prepared materials, when examined by X-ray diffraction, were found to crystallize in the rhombohedral $Th_2Zn_{17}$ structure (space group R $\bar{3}$ m). Magnetization measurements were performed on a SQUID magnetometer in the temperature range 5–300 K in magnetic fields up to 6 T. Magnetic measurements above 300 K were performed on a home-built Faraday balance.

The results of magnetic measurements showing the temperature dependence of the magnetization are displayed in Fig. 1. These results clearly demonstrate the

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0304-8853/98/$19.00 
*PII* S0304-8853(97)00827-5

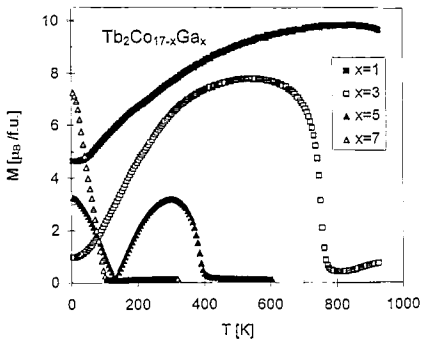


Fig. 1. Temperature dependence of the magnetization for $Tb_2Co_{17-x}Ga_x$ intermetallics measured in a field of 0.1 T.

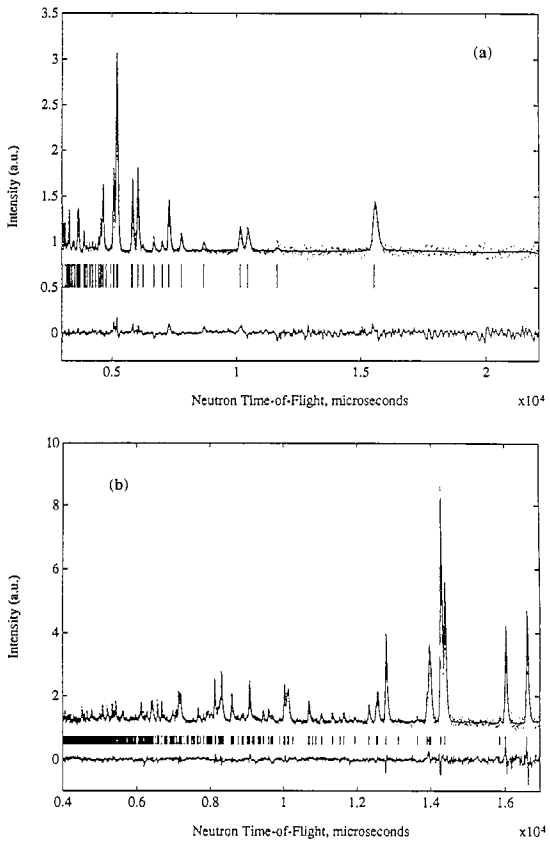


Fig. 2. Observed and calculated neutron-diffraction patterns for $Tb_2Co_{12}Ga_5$ at 293 K for (a) forward scattering and (b) backscattering detector banks.

ferrimagnetic coupling between Tb and Co sublattices with the compound for $x = 7$ demonstrating a compensation temperature at approximately 130 K. The neutron-diffraction data were analysed by the Rietveld method. Initially, a statistical occupancy was assumed for Co/Ga occupancies on the 6c [0, 0, $z$ ( $\sim 0.1$)], 9d [0, 0, 0], 18f [$x$( $\sim 0.29$), 0, 0] and 18h [$x$( $\sim 0.5$), $2x$, $z$( $\sim 0.15$)] sites. The large contrast in neutron-scattering lengths between Co and Ga means that the Co occupancies for the 6c, 9d, 18 and 18f sites can be determined with a good precision. Results of the refinements of the room temperature data are displayed in Table 1 whilst the observed and calculated neutron powder patterns for $Tb_2Co_{12}Ga_5$ at 293 K are displayed in Fig. 2. Also, displayed in Table 1 are some results of the bulk magnetization measurements, the Curie temperatures and the magnetization (per formula unit) at 5 K. Amongst the most prominent features displayed in Table 1 is the total avoidance of Ga atoms for the 9d sites, a result similar to that already observed for corresponding $Tb_2Fe_{17-x}Ga_x$ [5] and $Y_2Fe_{17-x}Ga_x$ compounds [6]. For $x > 3$, Ga atoms increasingly occupy the 6c and 18f sites, but at the cost of the 18h site, which shows a decreasing Ga occupancy for this concentration range. From the neutron-diffraction data, the compounds $Tb_2Co_{12}Ga_5$ and $Tb_2Co_{10}Ga_7$ are found to display planar and axial magnetic structures at 4.2 K, respectively. The largest Co moment is found at the 9d site, with 1.9(2) $\mu_B$ for $Tb_2Co_{16}Ga$ which is reduced to 0.8(2) $\mu_B$ for $Tb_2Co_{12}Ga_5$ at 293 K. The sign reversal of the Co sublattice anisotropy from planar to axial clearly occurs in a concentration range where Ga atoms substitute almost exclusively into the 18h site. The results also indicate a strong change in the crystal-field-induced Tb anisotropy with Ga concentration.

Table 1
Refined lattice parameters, Ga site occupancies (%), Curie points and saturation magnetization $M_s$ ($\mu_B$/formula unit at 4.2 K) for $Tb_2Co_{17-x}Ga_x$ intermetallics

| | $TbCo_{16}Ga$ | $TbCo_{14}Ga_3$ | $TbCo_{12}Ga_5$ | $TbCo_{10}Ga_7$ |
|---|---|---|---|---|
| $a$ (Å) | 8.3976 | 8.4591 | 8.5252 | 8.6519 |
| $c$ (Å) | 12.2384 | 12.3358 | 12.4088 | 12.3465 |
| Ga, 6c | 0.0 | 0.0 | 31.6(5) | 80.3(5) |
| Ga, 9d | 0.0 | 0.0 | 0.0 | 0.0 |
| Ga, 18f | 0.0 | 0.0 | 34.3(3) | 62.4(3) |
| Ga, 18h | 17.7(5) | 52.5(6) | 38.5(3) | 27.5(3) |
| $T_c$ (K) | > 900 | 755 | 388 | 95 |
| $M_s$ | 7.2 | 1.6 | 4.1 | 9.8 |

Note: Lattice parameters and occupancies are for 293 K.

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