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Physica B 211 (1995) 115–117

PHYSICA B

High-field magnetisation of RCo_9Si_2 compounds

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Abstract

We prepared RCo_9Si_2 compounds in polycrystalline form with R representing Sm, Gd, and Tb. The magnetisation in fields up to 35 T was studied at 4.2 K on powder particles free to orient in the applied magnetic field. The Sm compound is ferromagnetic, the Gd and Tb compounds were found to order ferrimagnetically. The moments on the Co-sublattice are found to be about $0.9\mu_B$/Co-atom. For the ferrimagnetic samples, the exchange-coupling constant J_{RT}/k can be estimated from the slope of the high-field magnetic isotherm. This coupling constant is found to be 6.8 K for both the Gd and the Tb compound. To achieve a proper description of the magnetisation of $TbCo_9Si_2$ a three-sublattice model is used.

1. Introduction

In the search for new hard-magnetic materials, information on the coupling between the rare-earth (R) and the transition metal (T) sublattice is important. By means of high-field magnetisation measurements on free single-crystalline powder particles this coupling can be derived for ferrimagnetic materials [1]. Here, we present results of high-field magnetisation measurements on compounds of the type RCo_9Si_2 with R representing Sm, Gd, Tb. These compounds order around 450 K, either ferro- or ferrimagnetically when formed with light- or heavy-R elements, respectively [2].

2. Experimental

We have prepared polycrystalline ingots of the composition RCo_9Si_2 with R representing Sm, Gd, Tb, Dy, Ho, Er, Tm and Lu, by arc melting metals of purity 99.85% or better in a water-cooled copper boat. Melting was done in a continuously Ti-gettered argon atmosphere. X-ray and electron-microprobe analysis showed as the as-cast ingots not to form in the desired $BaCd_{11}$ structure [3]. Subsequent annealing at 1050°C for 4 weeks led to almost single-phase samples for Sm, Gd, and Tb. The lattice parameters and the phase purity are summarised in Table 1. The lattice parameters are in good agreement with those of Ref. [3], but the crystal structure of the compounds with Gd and Tb was found to be of the $CeMn_6Ni_5$ type (space group 127), which is also reported for compounds of type RCo_9Ti_2 [4]. The compounds containing Dy, Ho, Er, Tm and Lu, were found to adopt the $ThMn_{12}$-type of structure and will not be treated here.

The samples were powdered under argon gas in a ball-milling machine and sieved (mesh 200) to ensure an upper limit (74 μm) of the grain size. The powder particles, which can be assumed to be single crystalline, were put into teflon sample holders, being free to rotate into their minimum-energy direction with respect to the external field during the measurements. High-field magnetisation measurements up to 35 T were performed in the High Magnetic Field Installation at the University of

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SSDI 0921-4526(94)00959-7

Table 1
Structural and magnetic properties of RCo_9Si_2 compounds

Compound	a (Å)	b (Å)	Impurities (% Co)	T_c (K)	$n_{RT} \times 10^{-23}$ (T/Am^2)	J_{RT}/k (K)	M_{co} (μ_B)
$SmCo_9Si_2$	9.779	6.320	—	530	—	—	0.9
$GdCo_9Si_2$	9.759	6.308	< 1	480	9.9	6.8	0.9
$TbCo_9Si_2$	9.77	6.13	< 5	463[a]	6.6	6.8	0.9

[a] From Ref. [2].

Amsterdam [5]. The measurements were performed at 4.2 K, using stepwise field-time profiles.

Several samples were measured in a high-temperature Faraday balance to determine the Curie temperatures.

3. Results and discussion

The obtained magnetic isotherms are displayed in Fig. 1. $SmCo_9Si_2$ is a ferromagnet with a saturation magnetisation, obtained by extrapolation to infinite field, of about $9.3\mu_B$/f.u.

The magnetic isotherm of $GdCo_9Si_2$ can be divided into two linear parts. First, the magnetisation slowly increases up to a critical field B_c of about 11 T where a pronounced upward kink occurs. Extrapolation to zero field from the high-field region gives an intercept with the magnetisation axis at $0.3\mu_B$/f.u. Extrapolation to zero field from the low-field region after correction by subtracting the extrapolated value of $0.3\mu_B$/f.u. derived from the high-field region gives a spontaneous magnetisation equal to $0.9\mu_B$/f.u.

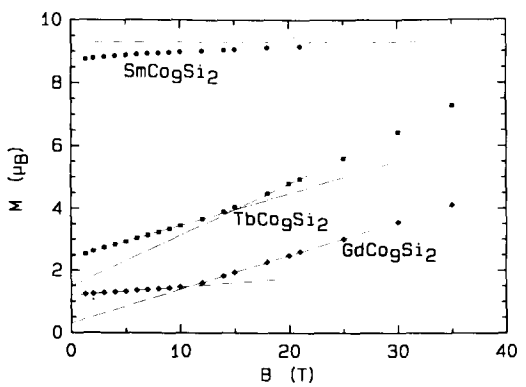

Fig. 1. Magnetic isotherms at 4.2 K for free powder samples of RCo_9Si_2 compounds (the double-dash–dotted line gives the saturation magnetisation of $SmCo_9Si_2$, the other lines represent least-squares fits to the data).

The magnetic isotherm of the Gd compound can be described in a simple two-sublattice model in which the compound is assumed to consist of one magnetic R sublattice and one magnetic T sublattice, which comprises both the Co and the Si atoms [1]. Assuming the free-ion value for the Gd moment and domination of the Co-sublattice moment we derive $7.9\mu_B$ for the Co-sublattice magnetisation and from this an average Co moment of $0.9\mu_B$. We then can identify the field B_c where the kink in the magnetisation of $GdCo_9Si_2$ occurs as the field where the Gd and the T sublattices depart from the antiparallel orientation. The high-field slope is inversely proportional to the inter-sublattice-molecular-field coefficient n_{RT}, which is related to the microscopic Heisenberg-type of coupling parameter J_{RT} by

$$n_{RT} = \frac{-J_{RT} Z_{RT}(g_R - 1)}{N_T \mu_B^2 g_R}, \tag{1}$$

where Z_{RT} is the number of T atoms surrounding an R atom, g_R the Landé factor, N_T the number of T atoms per formula unit and μ_B the Bohr magneton (in the $CeMn_6Ni_5$-structure type $Z_{RT} = 20$ and $N_T = 11$). A value of the coefficient J_{GdT}/k of 6.8K has been derived from a fit to the magnetic isotherms for $B > B_c$. The results are collected in Table 1.

For the $TbCo_9Si_2$ we find very similar behaviour as in the Gd isotype except that the low-field slope is much higher than for the Gd compound. The critical field is located at about 15 T. The magnetisation values extrapolated to zero field from the high-field region and from the low-field region are $1.5\mu_B$/f.u. and $2.4\mu_B$/f.u., respectively. Thus, we find for the spontaneous magnetisation a value equal to $0.9\mu_B$/f.u. The extrapolated value derived from the high-field region differs considerably from zero, which may be due to the presence of free Co which is confirmed by the microprobe analysis. A contribution of about 5 Vol% Co would account for the observed extrapolation value, which also agrees with the microprobe results (see Table 1).

The simple two-sublattice model seemingly does not hold for the Tb compound. The rather high initial slope found, might be attributed to misalignment of powder

particles but then one would expect a tendency towards saturation in the low-field part. A physical origin for the low-field slope would be bending of the moments of one sublattice. If Si atoms in the Co sublattice are assumed to be statistically distributed, they may generate some random variation of the crystal field at the Tb sites, leading to a non-collinear Tb sublattice. These rather weak fluctuations may be overcome by a comparatively low external field. For simplicity, we choose for the description of the magnetisation of the Tb compound a model including two non-collinear R sublattices with equal moments M_r, with a zero-field canting angle $2\phi_0$ [6]. In this description it is assumed that the two R sublattices have an effective inplane anisotropy constant K and the angle between the rare-earth sublattices 2ϕ is a new parameter in the energy expression which then becomes

$$E = C + 2n_{RT}M_rM_T\cos\alpha\cos\phi + 2K\sin 2\phi - B_0\sqrt{(2M_r\cos\phi)^2 + M_T^2 + 4M_rM_T\cos\alpha\cos\phi}, \quad (2)$$

where C is a constant anisotropy, α is the angle between the R- and T-sublattice moments, and B_0 the applied field. Analytical minimisation of this energy expression gives again a regime marked by a critical field B_c where the slope is inversely proportional to n_{RT}. This critical field is given by

$$B_c = n_{RT}|M_T - 2M_r\cos\phi_c|. \quad (3)$$

ϕ itself is field-dependent in the fields below the critical field which results in a non-zero slope. Under the assumption that also in this compound the T-sublattice moment dominates, and that the Co moment is equal to $0.9\mu_B$, the extremal values of ϕ obtained by fitting the experimental data are 36° and 50° in zero field and at B_c (15 T), respectively. For J_{TbT}/k we find 6.8 K and for K a value 5×10^{-22} J/f.u. is found.

In conclusion, compared with other RT compounds [1] we find rather low Co moments in the RCo_9Si_2 compounds and moderate values for the coupling constant J_{RT}. The non-collinearity of the R sublattice proposed for $TbCo_9Si_2$ is, however, open to discussion and should still be verified by microscopic tools as e.g. neutron diffraction.

Acknowledgements

This research was supported by the Technology Foundation (STW).

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