Vol. 92 (1997) ACTA PHYSICA POLONICA A No. 2

Proceedings of the European Conference "Physics of Magnetism 96", Poznań 1996

GROUND STATE PROPERTIES OF Tb^{3+} ION IN $TbNi_{10}Si_2$

P. STEFAŃSKI[+◇], W. SUSKI[*†], K. WOCHOWSKI[*] AND T. MYDLARZ[†]

[+]Institute of Molecular Physics, Polish Academy of Sciences
Smoluchowskiego 17, 60-187 Poznań, Poland
[◇]Institut für Festkörper und Werkstoffforschung Dresden e.V.
Postfach 270016, 01171 Dresden, Germany
[*]W. Trzebiatowski Institute of Low Temperature and Structure Research
Polish Academy of Sciences, P.O. Box 937, 50-950 Wrocław 2, Poland
[†]International Laboratory of High Magnetic Fields and Low Temperatures
Gajowicka 95, 50-539 Wrocław, Poland

Ground state properties of Tb^{3+} ion were investigated in the tetragonal $ThMn_{12}$-type $TbNi_{10}Si_2$ compound on the basis of temperature dependence of the paramagnetic susceptibility and magnetization measurements. The rare earth temperature dependence of the susceptibility was calculated using the Van Vleck equation. The $\Gamma_{t5}^{(1)}$ doublet is suggested to be a ground state separated by 9 K from the first excited singlet $\Gamma_{t1}^{(2)}$. The magnetic moment associated with the ground state doublet agrees with saturation magnetization data at 4.2 K. The overall crystal field splitting is estimated to be 105 K.

PACS numbers: 75.30.Gw, 75.50.Gg

The recent permanent magnet research stimulates investigations of various anisotropic iron-rich intermetallics. Among them there are compounds of the form $RFe_{12-x}T_x$ (T = Ti, V, Cr, Mo, W and Si [1]) with tetragonal $ThMn_{12}$-type structure, which are isomorphous with non-magnetic $RNi_{10}Si_2$ series. The paramagnetic behaviour of the latter compounds is dominated by the crystal-field effects in the rare earth subsystem, since the magnetic moment at Ni sites is not significant [2]. The aim of the present paper is to investigate the crystal-field level structure of Tb^{3+} with the use of magnetic measurements in $TbNi_{10}Si_2$. The Ni subsystem can be considered as a Pauli paramagnet as already observed in $YNi_{10}Si_2$ [3]. It is wort h noting that neutron spectroscopy measurements performed on a sample containing Tb [2] showed crystal-field level excitations that were not well resolved due to a complex level scheme.

The method of preparation of $TbNi_{10}Si_2$ and $YNi_{10}Si_2$ has been described elsewhere [4]. The alloyed samples have been annealed for three weeks at 1100°C. Magnetic susceptibility measurements were carried out in the temperature range

4.2–1000 K using the Faraday method. The corrections resulting from diamagnetic effects are taken into account. Magnetization measurements at 4.2 K have been performed with the use of an extraction type magnetometer in magnetic fields up to 14 T. The temperature dependence of the paramagnetic susceptibility of $TbNi_{10}Si_2$ (and of $YNi_{10}Si_2$ for comparison [3]) in a low temperature region is plotted in Fig. 1. The effective magnetic moment obtained from the susceptibility in the high temperature region, being $p_{\mathrm{eff}} = 9.67\mu_{\mathrm{B}}$, is very close to the calculated rare earth ion moment $g_J[J(J+1)]^{1/2} = 9.72\mu_{\mathrm{B}}$. It indicates that the magnetism in $TbNi_{10}Si_2$ is only due to the rare earth free ions. The paramagnetic Curie point Θ, extrapolated from susceptibility measurements down to liquid helium temperature, is at 0 K. The field dependence of the magnetization exhibits a paramagnetic saturation, on which a slight increase with the field due to the induced Ni moments is superimposed. The magnetization in the saturation state is $5.0\mu_{\mathrm{B}}$/f.u.

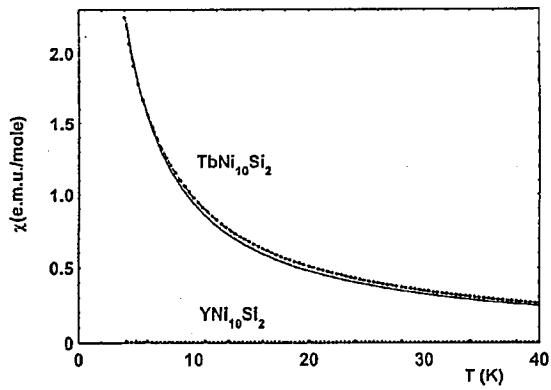

Fig. 1. Temperature dependences of the susceptibility of $TbNi_{10}Si_2$ and $YNi_{10}Si_2$. The solid line represents the fitted theoretical curve to the temperature dependence of the magnetic susceptibility of $TbNi_{10}Si_2$.

In the $TbNi_{10}Si_2$ compound of $ThMn_{12}$-type structure the Tb^{3+} ion occupies the crystallographic 2(a) site of the D_{4h} tetragonal symmetry. The crystal electric field (CEF) Hamiltonian for such a symmetry has a form

$$H_{\mathrm{CEF}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4 + B_6^0 O_6^0 + B_6^4 O_6^4, \tag{1}$$

where B_n^m are CEF parameters and O_n^m are Stevens operators [5].

The ground state multiplet $^7F_6$ of the Tb^{3+} free ion is split by the crystal field potential of tetragonal symmetry (Eq. (1)) according to the following decomposition:

$$D^6 = 2\Gamma_{t1} + \Gamma_{t2} + 2\Gamma_{t3} + 2\Gamma_{t4}3 + \Gamma_{t5}, \tag{2}$$

where Γ_{tn}, $n = 1, 2, 3, 4$ are singlets and Γ_{t5} is a doublet.

Using the set of crystal field functions as a basis, the CEF Hamiltonian has been reduced to the block-diagonal form, each block corresponding to an appropriate irreducible representation Γ_{tn}. The rare earth ion susceptibility has been

calculated using the Van Vleck formula for $\chi_{\parallel}$ (along [001] direction) and $\chi_{\perp}$. The total rare earth ion susceptibility χ_R for polycrystalline sample of tetragonal symmetry with randomly oriented grains has been obtained from the following relation:

$$\chi_R = \frac{1}{3}\chi_{\parallel} + \frac{2}{3}\chi_{\perp}. \tag{3}$$

The Ni contribution to the total susceptibility has been considered to be of a Pauli paramagnetic type. The approach proposed in Ref. [6] leads to the following expression for the temperature dependence of the total susceptibility:

$$\chi_T(T) = \chi_{Ni} + (1+\alpha)^2\chi_R(T), \tag{4}$$

where χ_{Ni} — Pauli susceptibility of Ni, χ_R — Van Vleck susceptibility of the rare earth ion and $\alpha = n_{RNi}\chi_{Ni}$ n_{RNi} is R–Ni exchange coefficient. The above formula has been used in the fit procedure in order to obtain the B_n^m crystal field parameters. The numerical fit to the experimental data has been based on the so-called optimization simplex method [7]. For calculation of $\chi_R(T)$, as the first step the B_n^m parameters have been taken from Ref. [8] for isostructural $DyFe_{10}Cr_2$ compound, and then each block corresponding to an appropriate irreducible representation Γ_{tn} has been diagonalized separately. The total susceptibility including Ni contribution has been calculated from Eq. (4). The susceptibility of Ni has been taken $\chi_{Ni} = 0.013$ [e.m.u./mole] [3] and kept constant in the whole range of temperatures. The best fit to the experimental data is shown in Fig. 1 (solid line) and the obtained CEF parameters are: $B_2^0 = 0.34(1)$ K, $B_4^0 = 0.63(1)$ mK, $B_4^4 = 1.31(1)$ mK, $B_6^0 = -0.12(1)$ mK and $B_6^4 = 0.13(1)$ mK. The exchange coefficient between the rare earth and nickel is calculated to be $n_{RNi} = 158$ [mole/e.m.u.]. The value of $A_2^0 = -41$ K$/a_0^2$ (a_0 — a Bohr radius), calculated from the relation $B_2^0 = \alpha_J\langle r^2\rangle A_2^0$, (where α_J — the second order Stevens factor [5] and $\langle r^2\rangle$ — radial integral for the $4f$ electrons [9]), is comparable in magnitude with $A_2^0 = -60$ K$/a_0^2$ given from Mössbauer data for an isostructural compound $GdFe_{10}Si_2$ [10], with an assumption of shielding factor $\sigma_2 = 0.5$.

TABLE

The calculated crystal field energy levels of Tb^{3+} ion in $TbNi_{10}Si_2$.

Irreducible representation	$\Gamma_{t5}^{(1)}$	$\Gamma_{t1}^{(2)}$	$\Gamma_{t3}^{(1)}$	Γ_{t2}	$\Gamma_{t4}^{(2)}$	$\Gamma_{t1}^{(1)}$	$\Gamma_{t3}^{(2)}$	$\Gamma_{t5}^{(3)}$	$\Gamma_{t4}^{(1)}$	$\Gamma_{t5}^{(2)}$
Energy in [K]	0	9.4	21.6	37.3	39.0	44.0	47.1	53.0	73.7	105.0

The energy level sequence obtained from the fit procedure is presented in Table. The ground level of Tb^{3+} is a doublet $\Gamma_{t5}^{(1)}$ and the first excited level is a singlet $\Gamma_{t1}^{(2)}$, about 9 K above the ground state. The overall crystal field splitting is of the order of 100 K, which is comparable in magnitude with overall crystal field splitting for Er^{3+} ion (about 70 K) obtained by inelastic neutron scattering measurements [2]. The calculated magnetic moment associated with the ground state doublet is equal to $4.56\mu_B$/Tb ion. This is an evidence of partial

quenching of the free rare earth ion moment by the crystal field. The calculated value is lower than the one obtained experimentally from the field dependence of the magnetization ($5.0\mu_B$/f.u.). However, the induced Ni moment increases linearly with the field and for $H_{app} = 140$ kOe it has a value of $0.33\mu_B$/f.u. in $YNi_{10}Si_2$. It is aligned parallel to the external field and the rare earth magnetic moment, leading to the total magnetic moment of $4.89\mu_B$/f.u. in $TbNi_{10}Si_2$, which is comparable to the experimental value.

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