Magnetic coupling in rare earth–nickel compounds of the type R_2Ni_7

G. F. Zhou and F. R. de Boer

Van der Waals-Zeeman Laboratory, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam (Netherlands)

K. H. J. Buschow Philips Research Laboratories, P.O. Box 80.000, 5600 JA Eindhoven (Netherlands)

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Abstract

The field dependence (up to 35 T) of the magnetization at 4.2 K of several compounds of the type $R_{2-x}Y_xNi_7$ was studied. The magnetic isotherms at 4.2 K were analysed with a mean-field model, from which the magnetic-coupling constant J_{RNI} between the moments of the rare earths and nickel in the Hamiltonian $H = \sum_R J_{RNI} 2S_R \cdot S_{NI}$ was estimated. It was found that the coupling constants in the R_2Ni_7 compounds are almost twice as large as in the R_2Ni_{17} compounds studied previously.

1. Introduction

In a number of preceding investigations [1-3], we have studied ferrimagnetic rare earth (R)-transition metal compounds in high magnetic fields. Profiting from the possibility of being able to rotate the two antiparallel sublattice magnetizations towards each other, we have determined the corresponding intersublattice-coupling constants [4]. In the present study, we have extended our investigation to R–Ni compounds of the type R₂Ni₇, with the purpose of determining how far these coupling constants (when expressed per pair of unit spins) depend on the 3d moment and/or on the 3d atom concentration. In contrast with the many R–Co and R–Fe compounds, there are relatively few R–Ni compounds in which the nickel sublattice is magnetic. This is, for instance, the case in compounds of the type R₂Ni₁₇ studied before. The nickel sublattice magnetization is zero in compounds of the type RNi₅ but owing to 5d–3d hybridization effects the nickel sublattice again acquires a small moment in compounds of the type R₂Ni₇ studied in the present investigation [5].

2. Experimental details

Various compounds of the type R_2Ni_7 were prepared by arc melting, followed by vacuum annealing at 1050 °C for 3 weeks. It was confirmed by

X-ray diffraction measurements that all samples prepared in this way were approximately single phase and of the rhombohedral Gd_2Co_7 -type structure.

Magnetic isotherms at 4.2 K of these compounds were measured in the high field installation at the University of Amsterdam [6] in fields up to 35 T. The samples were in the form of finely powdered material, so that the individual particles were free to rotate into their minimum-energy direction during the measurements.

Representative examples of magnetic measurements are shown in Figs. 1–3. In most cases the magnetization at 4.2 K was measured at discrete values of the field strength. For the two compounds $Gd_{2-x}Y_xNi_7$ with x = 1.80 and 1.85, data were also taken during a field pulse in which the field decreased linearly in time from 38 T at a rate of 55 T s⁻¹. The results of the latter measurements are represented by the two full curves in Fig. 1.



Fig. 1. Field dependence of the magnetic moment in various $Gd_{2-x}Y_xNi_7$ compounds at 4.2 K obtained on fine powder particles free to rotate in the field applied.



Fig. 2. Field dependence of the magnetic moment in various $Tb_{2-x}Y_xNi_7$ compounds at 4.2 K obtained on fine powder particles free to rotate in the field applied.



Fig. 3. Field dependence of the magnetic moment in various $Ho_{2-x}Y_xNi_7$ compounds at 4.2 K obtained on fine powder particles free to rotate in the field applied.

3. Discussion

As was done before, we shall analyse our data by means of a twosublattice mean-field model [1-4]. For convenience we repeat here the ideal behaviour that may be expected on the basis of this model for compounds in which the rare earth sublattice magnetization $M_{\rm R}$ and 3d sublattice magnetization $M_{\rm T}$ are sufficiently close to each other in value. In comparatively low fields, the moment configuration is strictly antiparallel and the magnetization is equal to the values $M_1 = |M_{\rm R} - M_{\rm Ni}|$. Beyond a critical field strength $(B_1^{\rm crit} = |M_{\rm R} - M_{\rm Ni}| n_{\rm RNi})$, the exactly antiparallel moments start to bend towards each other and the magnetic moment is described by $M = B/n_{\rm RNi}$ and thus

$$\frac{\mathrm{d}M}{\mathrm{d}B} = n_{\mathrm{RNi}}^{-1} \tag{1}$$

The parameter $n_{\rm RNi}$ can therefore be derived straightforwardly from the high field slopes obtained for $B > B_1^{\rm crit}$. At higher fields, beyond a critical value $B_2^{\rm crit} = |M_{\rm R} + M_{\rm Ni}| n_{\rm RNi}$, the forced ferromagnetic alignment of the magnetic moments is obtained, corresponding to a magnetization equal to $M_2 = M_{\rm R} + M_{\rm Ni}$.

The saturation moment M_1 derived from the data displayed for $\mathrm{Gd}_2\mathrm{Ni}_7$ in Fig. 1 by extrapolation to B=0 equals $M_s=12.8\mu_{\mathrm{B}}$ (f.u.)⁻¹, where f.u. is a formula unit. With $M_{\mathrm{Gd}}=14\mu_{\mathrm{B}}$ (f.u.)⁻¹ this leaves $1.2\mu_{\mathrm{B}}$ (f.u.)⁻¹ for M_{Ni} . A similarly large difference between M_{R} and M_{Ni} is found for the other $R_2\mathrm{Ni}_7$ compounds, meaning that the values of $|M_{\mathrm{R}}-M_{\mathrm{Ni}}|$ are too large for B_1^{crit} to fall into the field range accessible to us. Only for the compounds with $x \ge 1.8$, does the difference in intrasublattice magnetization become sufficiently small to give rise to the behaviour needed for the determination of n_{RNi} via eqn. (1). This is the reason why we considered mainly yttrium-rich compounds in this investigation. The values of n_{RNi} and M_{Ni} have been listed in Table 1. For $\mathrm{Gd}_2\mathrm{Ni}_7$ and $\mathrm{Gd}_{0.2}\mathrm{Y}_{1.8}\mathrm{Ni}_7$, the nickel sublattice moments M_{Ni} have been derived from $M_1 = |M_{\mathrm{R}} - M_{\mathrm{Ni}}|$. For all the other compounds, M_{Ni} has been derived from $M_2 = M_{\mathrm{R}} + M_{\mathrm{Ni}}$.

TABLE 1

Intersublattice magnetic-coupling constants $n_{\rm RNi}$ and exchange-coupling constants $J_{\rm RNi}/k$ for $R_{2-x}Y_xNi_7$ compounds, where the nickel sublattice moments $M_{\rm Ni}$ were derived from the corresponding saturation moments $|M_{\rm R}-M_{\rm Ni}|$, or from $M_{\rm R}+M_{\rm Ni}$ (see main text), using values for the R moments as listed under the heading $\mu_{\rm R}$

Compound	$n_{\rm RNi}$ (T (f.u.) ⁻¹ $\mu_{\rm B}^{-1}$)	J _{RNi} /k (K)	$\mu_{ m R}$ ($\mu_{ m B}$ atom ⁻¹)	$\begin{array}{l} M_{\rm Ni} \\ (\mu_{\rm B} \ ({\rm f.u.})^{-1}) \end{array}$
Gd _{0.08} Y _{1.92} Ni ₇	24.12	- 15.1	(7)	0.83
$Gd_{0,1}Y_{1,9}Ni_7$	17.8	-11.2	(7)	0.42
Gd _{0.15} Y _{1.85} Ni ₇	17.2	-10.8	(7)	0.69
Gd _{0.2} Y _{1.8} Ni ₇	24.5	-15.4	(7)	0.56
Gd ₂ Ni ₇	-		(7)	1.2
Tb _{0.1} Y _{1.9} Ni ₇	12.1	-11.4	(9)	0.45
Tb _{0 15} Y _{1 85} Ni ₇	10.3	-9.7	(9)	0.55
$Dy_{0,1}Y_{1,9}Ni_7$		-	(10)	0.09
$Dy_{0.14}Y_{1.86}Ni_7$	8.4	-10.6	(10)	0.52
$H_{0,1}Y_{1,9}Ni_7$	5.4	-8.4	(10)	0.50
Ho _{0 15} Y _{1.85} Ni ₇	4.8	-7.6	(10)	0.62
$Er_{0,1}Y_{1,9}Ni_7$	4.7	8.8	(9)	0.63
Er _{0.15} Y _{1.85} Ni ₇	4.8	-9.1	(9)	0.47
Y ₂ Ni ₇		-	-	0.55



Fig. 4. Dependence of the coupling constant J_{RNi} in R_2Ni_{17} and R_2Ni_7 compounds on the R component. For the R_2Ni_7 compounds the average values from Table 1 are plotted. For the R_2Ni_{17} compounds we used the data from ref. 7.

The various n_{RNi} values listed in Table 1 have been transformed into coupling constants J_{RNi} per pair of unit spins as defined by the exchange Hamiltonian $H_{\text{exch}} = \Sigma 2 J_{\text{RNi}} S_{\text{R}} S_{\text{Ni}}$ via the expression

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$$n_{\rm RNi} = \frac{-J_{\rm RNi} Z_{\rm RNi} (g_{\rm R} - 1)}{N_{\rm Ni} \mu_{\rm B}^2 g_{\rm R}} \tag{2}$$

In the crystal structure of the R_2Ni_7 compounds, the average number Z_{RNi} of nearest transition metal atom neighbours to an R atom equals 15. The number N_{Ni} of transition metal atoms per formula unit equals 7. It is seen from Table 1 and also from Fig. 4 that there is a tendency of $|J_{RNi}|$ to decrease from $R \equiv Gd$ towards the end of the lanthanide series. We have included in Fig. 4 the J_{RNi} values derived by us previously for R_2Ni_{17} compounds [7]. It may be seen that the absolute values of the coupling constants J_{RNi} determined for the R_2Ni_7 compounds in the course of the present investigation are substantially larger than the corresponding values in the R_2Ni_{17} series. A similar tendency of the J_{RT} coupling constant to increase with decreasing transition metal T concentration was observed also for R–Fe and R–Co compounds.

4. Conclusion

The results obtained in the course of the present investigation have shown that the concentration dependence of the magnetic coupling constant $J_{\rm RT}$ follows the same general behaviour in R–T compounds. Independent of whether T represents iron, cobalt or nickel, there is a strong tendency for $J_{\rm RT}$ to increase with decreasing T concentration. Apparently this tendency is not very dependent on the moment value of the T component, the nickel moments in R₂Ni₇ being more than an order of magnitude smaller than the cobalt moments in R₂Co₇ [3].

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