ELSEVIER

Journal of Alloys and Compounds 239 (1996) 147–149

Journal of ALLOYS AND COMPOUNDS

Magnetic properties of $R_6Fe_{11}Al_3$ and $R_6Fe_{12}Al_2$ compounds with R = Pr, Nd

Z.G. Zhao, F.R. de Boer, K.H.J. Buschow*

Van der Waals–Zeeman Institute, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, Netherlands

Received 9 January 1996

Abstract

The magnetic properties of $R_6Fe_{11}Al_3$ and $R_6Fe_{12}Al_2$ compounds with R = Pr and Nd have been investigated by measuring the magnetization of free powder particles at 4.2 K in high magnetic fields. Metamagnetic transitions were found in all four compounds. The transition fields and the saturation magnetization decrease with increasing Al content for both the Pr and the Nd compounds.

Keywords: Ternary rare earth compounds; R–Fe–Al compounds; Magnetic properties; Metamagnetism; High-field measurements

1. Introduction

The compounds $R_6Fe_{11}Al_3$ and $R_6Fe_{12}Al_2$ with R = Pr and Nd crystallize in the $La_6Co_{11}Ga_3$-type structure with space group *I4/mcm* [1]. In these compounds, La has two different crystallographic sites. In the search for novel Fe-based permanent-magnet materials, such compounds have been studied by several groups in the last few years [2–11]. For Fe-based compounds, the $La_6Co_{11}Ga_3$-type structure is stable only for a few light-rare-earth elements (La, Pr, Nd and Sm) [1]. Owing to the different local environment of each different crystallographic site, complex magnetic properties can be expected. Our earlier results have shown that in these compounds antiferromagnetic coupling may exists between light-rare-earth (Pr and Nd) and iron moments [9]. This antiferromagnetic coupling strength is very sensitive to the nature and concentration of the third component, such as Al, Ga or B. As it has been explained by Campbell that antiferromagnetic coupling can only be expected between the moments of heavy-rare-earth and transition-metal ions in intermetallic compounds [12], it is important to investigate the origin of this antiferromagnetic coupling in this type of compound. Another interesting feature of this type of compound is that upon substitution of B for Ga, the magnetic anisotropy in the Nd-based compounds changes from easy-plane-type at low temperature to easy-axis at room temperature [7]. Furthermore, the Curie temperature is increased. In the present paper, the influence of Al on the high field magnetic properties of Pr- and Nd-based compounds with the $La_6Co_{11}Ga_3$-type structure is studied.

* Corresponding author.

2. Experimental

Alloys of $R_6Fe_{11}Al_3$ and $R_6Fe_{12}Al_2$ with R = Pr and Nd were prepared by arc melting together appropriate amounts of the pure elements in an atmosphere of very pure Ar. The ingots were annealed at 600°C for 6 weeks in an ampoule filled with pure Ar, followed by quenching in ice water. X-ray diffraction results showed only a negligible amount of impurity phases present.

The magnetization was measured at 4.2 K in high magnetic fields in the High-Field Facility at the University of Amsterdam. These measurements were carried out on fine powder particles that could rotate freely in the sample holder and hence could be orientated into their minimum energy configuration by the external field.

PII S0925-8388(96)02203-7

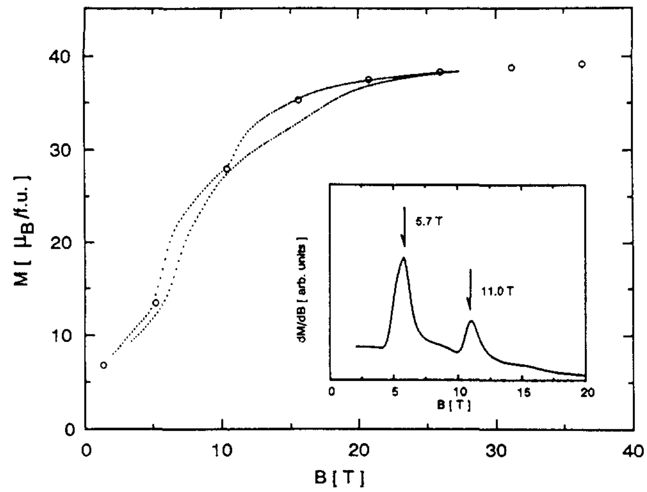

Fig. 1. High-field magnetization of $Nd_6Fe_{12}Al_2$ measured at 4.2 K on a free-powder sample. The dotted lines correspond to data taken in magnetic fields increasing and decreasing linearly with time. The circles stand for the measurements done in fields that vary in a step-wise way. The insert shows dM/dB for the measurement in decreasing field.

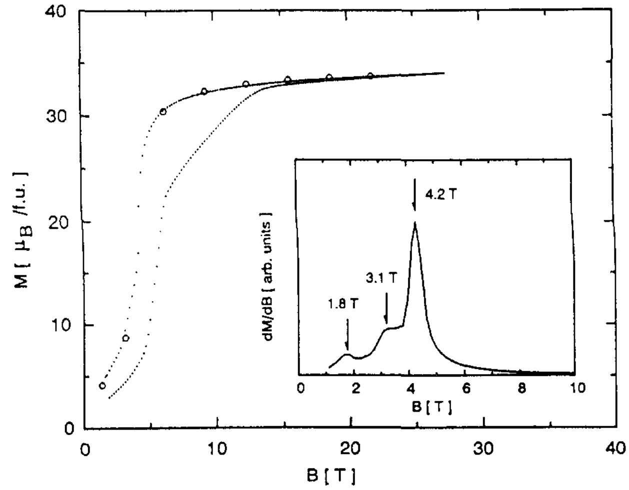

Fig. 3. High-field magnetization of $Pr_6Fe_{12}Al_2$ measured at 4.2 K on a free-powder sample. The dotted lines correspond to data taken in magnetic fields increasing and decreasing linearly with time. The circles stand for the measurements done in fields that vary in a step-wise way. The insert shows dM/dB for the measurement in decreasing field.

3. Results and discussion

The free-powder magnetization curves of the compounds are shown in Figs. 1 to 4. In order to show the magnetic transitions more clearly, we have plotted dM/dB as a function of the external field for the curves measured in decreasing field. All compounds, except $Pr_6Fe_{11}Al_3$, show two clear metamagnetic transitions. $Pr_6Fe_{11}Al_3$ even shows three transitions. We have shown previously for the $Nd_{6-x}La_xFe_{12}Ga_2$ system, that magnetic dilution of the rare-earth sublattice tends to increase the critical field of the magnetic phase transitions [10]. The present results show that the effect of magnetic dilution of the Fe sublattice has the opposite effect. As may be seen from Table 1, increasing substitution of Al for Fe leads to lower values of the lower as well as of the higher critical fields. This behaviour is further illustrated in Fig. 5.

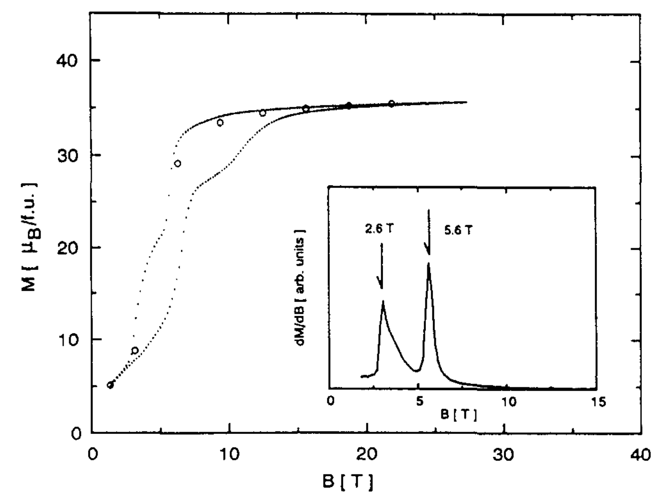

Fig. 2. High-field magnetization of $Nd_6Fe_{11}Al_3$ measured at 4.2 K on a free-powder sample. The dotted lines correspond to data taken in magnetic fields increasing and decreasing linearly with time. The circles stand for the measurements done in fields that vary in a step-wise way. The insert shows dM/dB for the measurement in decreasing field.

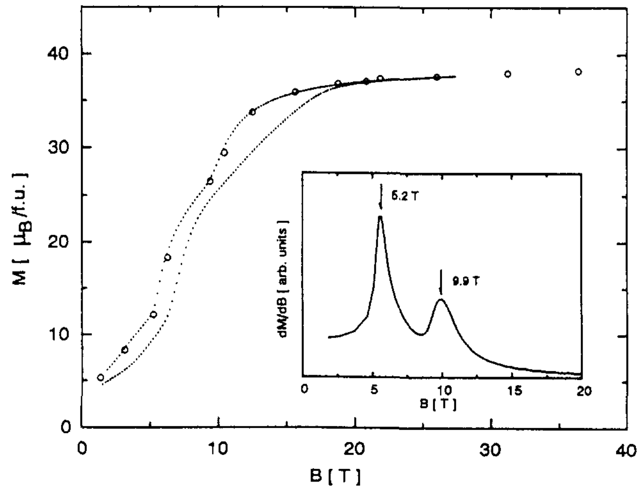

Fig. 4. High-field magnetization of $Pr_6Fe_{11}Al_3$ measured at 4.2 K on a free-powder sample. The dotted lines correspond to data taken in magnetic fields increasing and decreasing linearly with time. The circles stand for the measurements done in fields that vary in a step-wise way. The insert shows dM/dB for the measurement in decreasing field.

From the fact that the magnetization almost vanishes in zero applied field it can be derived that the rare-earth sublattice as well as the iron sublattice do not contribute much to the overall magnetisation in zero field. This probably means that the magnetic structure of each of the two sublattices is antiferro-

Table 1
Magnetic properties of $R_6Fe_{11}Al_3$ and $R_6Fe_{12}Al_2$ (R = Pr and Nd) compounds at 4.2 K

	M_s (μ_B/f.u.)	M_R (μ_B/R)	M_T (μ_B/Fe)	B_{tr}^{low} (T)	B_{tr}^{high} (T)
$Pr_6Fe_{12}Al_2$	38.5	3.20	1.61	5.2	9.9
$Pr_6Fe_{11}Al_3$	35.2	3.20	1.45	1.8	4.2
$Nd_6Fe_{12}Al_2$	39.5	3.27	1.66	5.7	11.0
$Nd_6Fe_{11}Al_3$	36.4	3.27	1.53	2.6	5.6

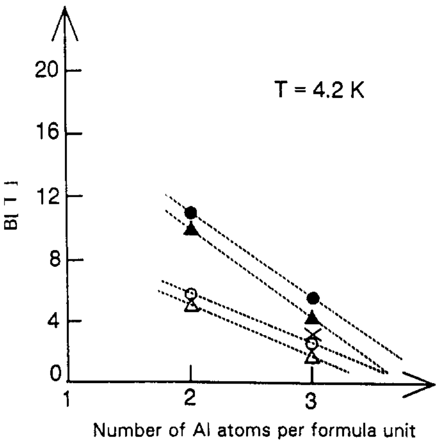

Fig. 5. Dependence of the transition fields in $Nd_6Fe_{12}Al_2$ and $Nd_6Fe_{11}Al_3$: (○: B_{tr}^{low} and ●: B_{tr}^{high}); $Pr_6Fe_{12}Al_2$ and $Pr_6Fe_{11}Al_3$ (△: B_{tr}^{low} and ▲: B_{tr}^{high}) and $Pr_6Fe_{11}Al_3$ (×) on the Al content.

magnetic or even more complex. The results of the high-field measurements displayed in Figs. 1–4 show that the compensating moment arrangements are broken in two or even more steps.

Values for the saturation magnetization of the compounds have been derived from the curves measured in decreasing field by extrapolation to very high fields. The results are shown in Table 1. The magnetic moments per Fe ion have been derived from the total magnetic moments by assuming the rare-earth moments to have the free-ion values. The Fe moments fall into the range 1.6–1.7 μ_B for the compounds with composition $R_6Fe_{12}Al_2$. This is almost equal to the Fe moments derived previously by the same procedure for the corresponding $R_6Fe_{12}Ga_2$ compounds [10].

It can also be seen in Table 1 that the magnetic moment of the Fe sublattice decreases with increasing Al content. In the molecular-field approximation, the magnetic coupling energy between two antiparallel coupled sublattices is proportional to the product of the magnetic moments of the two sublattices considered. The fact that both the critical fields and the Fe moments decrease with increasing Al content can therefore be taken as an indication that the Fe sublattice is involved in both types of magnetic phase transition. However, it should be borne in mind that there are five crystallographically non-equivalent Fe sites and two rare-earth sites in this type of compound, which may be accompanied by a variety of types of magnetic coupling between the different sublattices, all with different strengths. A more detailed description of the magnetic behaviour of the compounds studied in the course of the present investigation can therefore only be given after more experimental information has become available, including neutron-diffraction experiments.

References

[1] Yu.B. Kuzma and N.S. Bilonizhko, *Kristallographia, 18* (1973) 710.
[2] F. Weitzer and P. Rogl, *J. Less-Common Met., 167* (1990) 135.
[3] H. Klesnar and P. Rogl, *J. Mater. Res., 6* (1991) 53.
[4] O.M. Sichevich, R.V. Lapunova, A.N. Sobolev, Uu.N. Grin and Ya.P. Yarmolyuk, *Sov. Phys. Crystallogr., 30* (1985) 627.
[5] B.P. Hu, J.M.D. Coey, H. Klesnar and P. Rogl, *J. Magn. Magn. Mater., 117* (1992) 225.
[6] H.S. Li, B.P. Ho, J.M. Gadogan, J.M.D. Coey and J.P. Gavigan, *J. Appl. Phys., 67* (1990) 4841.
[7] Z.G. Zhao, X.K. Sun, Y.C. Chuang and F.R. de Boer, *J. Alloys Comp., 190* (1993) 72.
[8] F. Weitzer, A. Leithe-Jasper, P. Rogl, K. Hiebl, H. Noel, G. Wiesinger and W. Steiner, *J. Solid State Chem., 104* (1993) 368.
[9] Z.G. Zhao, F.R. de Boer, V.H.M. Duijn, K.H.J. Buschow and Y.C. Chuang, *J. Appl. Phys., 75* (1994) 7117.
[10] G. Zhao, C.H. de Groot F.R. de Boer and K.H.J. Buschow, *Physica B, 211* (1995) 102.
[11] C.H. de Groot, F.R. de Boer, K.H.J. Buschow, D. Hautot, G.J. Long and F. Grandjean, *J. Alloys Comp., 233* (1996) 161.
[12] I.A. Campbell, *J. Phys. F:, 2* (1972) L47.