Magnetic and Mössbauer spectral properties of the compound Nd$_6$Fe$_{13}$Au

C.H. de Groot[a,b], F.R. de Boer[a], K.H.J. Buschow[a], Dimitri Hautot[c], Gary J. Long[c],
F. Grandjean[d]

[a]*Van der Waals-Zeeman Institute, University of Amsterdam, Valckenierstraat 65, NL-1018 XE Amsterdam, Netherlands*
[b]*Philips Research Laboratories, Nl-5600 JA Eindhoven, Netherlands*
[c]*Department of Chemistry, University of Missouri-Rolla, Rolla, MO 65401-0249, USA*
[d]*Institute of Physics B5, University of Liège, B-4000 Liège, Belgium*

Abstract

The magnetic properties of Nd$_6$Fe$_{13}$Au were studied by means of magnetic measurements and iron-57 Mössbauer spectroscopy. It is shown that magnetic ordering of the Fe moments occurs at temperatures much above room temperature (T_N = 408 K). At low temperatures, the net moment of Nd$_6$Fe$_{13}$Au is very small owing to mutual compensation of the contributions of the various magnetic sublattices involved. High-field measurements made at 4.2 K show that ferromagnetic alignment of the sublattice moments is reached in two steps via first-order magnetic phase transitions. The Mössbauer spectra reveal that the Zeeman splitting gradually decreases with increasing temperature and that the average hyperfine field is zero at 435 K and above. Between 325 K and 411 K the area in the Mössbauer spectra due to the ordered magnetic component decreases gradually as it is replaced by a quadrupole doublet.

Keywords: Ternary rare earth compounds; Nd$_6$Fe$_{13}$Au; Magnetic properties; Mössbauer spectroscopy

1. Introduction

The compound Nd$_6$Fe$_{13}$Au crystallises in the tetragonal Nd$_6$Fe$_{13}$Si structure [1], which is an ordered variant of the La$_6$Co$_{11}$Ga$_3$ structure [2]. The magnetic properties of this compound were studied earlier by Weitzer et al. [3]. According to the latter authors the compound Nd$_6$Fe$_{13}$Au orders magnetically below T_C = 340 K, the magnetic ordering being accompanied by a strong rise of the magnetisation below T_C. Our recent measurements on the same compound have indicated quite different magnetic behaviour, a behaviour characterised by a Néel-type transition at a much higher temperature. This difference in magnetic behaviour has led us to study the magnetic properties of this compound in more detail.

2. Experimental

A sample of Nd$_6$Fe$_{13}$Au was prepared by arc melting starting materials of at least 99.9% purity. After arc melting the sample was wrapped in Ta foil, sealed in an evacuated quartz tube, annealed for 4 weeks at 600°C and quenched to room temperature. The X-ray diffraction diagrams showed that the annealed sample was approximately single phase and that its crystal structure corresponded to the tetragonal Nd$_6$Fe$_{13}$Si structure type reported by Allemand et al. [1].

The Mössbauer spectra were obtained on a constant-acceleration spectrometer which utilised a room temperature rhodium matrix cobalt-57 source and was calibrated at room temperature with α-Fe foil. The Mössbauer absorbers, which were typically 30 mg cm^{-2}, were prepared from pieces of samples pulverised under liquid toluene to particle diameters of approximately 0.045 mm or smaller. The Mössbauer spectra above room temperature were measured under vacuum. Details of the spectra analysis can be found elsewhere [4].

The field-dependence of the magnetisation of

$Nd_6Fe_{13}Au$ was measured in the high-field installation at the University of Amsterdam [5]. The measurements were performed on free powders in quasi-stationary measuring fields, as well as in fields increasing or decreasing linearly as a function of time.

Complementary magnetic measurements were made on a SQUID magnetometer in the temperature range 5–300 K in magnetic fields up to 5.5 T, whereas above 300 K the magnetic measurements were made on a home-built magnetometer based on the Faraday principle.

3. Results and discussion

Results of the magnetic measurements are displayed in Fig. 1. The prominent feature in the temperature dependence of the magnetisation is the occurrence of a moderately sharp cusp-like anomaly at $T_N = 408$ K. Below about 340 K there is a comparatively strong increase of the magnetisation, reminescent of the results reported previously by Weitzer et al. [3]. The value of the magnetisation remains fairly low in the entire temperature range considered, approximately 1 μ_B per formula unit of $Nd_6Fe_{13}Au$. This indicates that neither the Fe sublattice nor the Nd sublattice gives rise to a ferromagnetic moment arrangement in the temperature range studied.

Results of the high-field measurements at 4.2 K are shown in Fig. 2. Extrapolation of the low-field part of
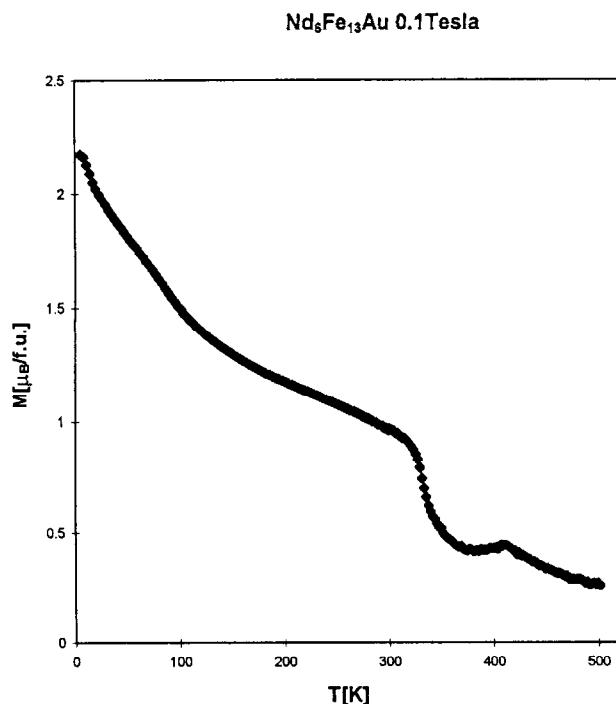

Nd$_6$Fe$_{13}$Au at 4.2K
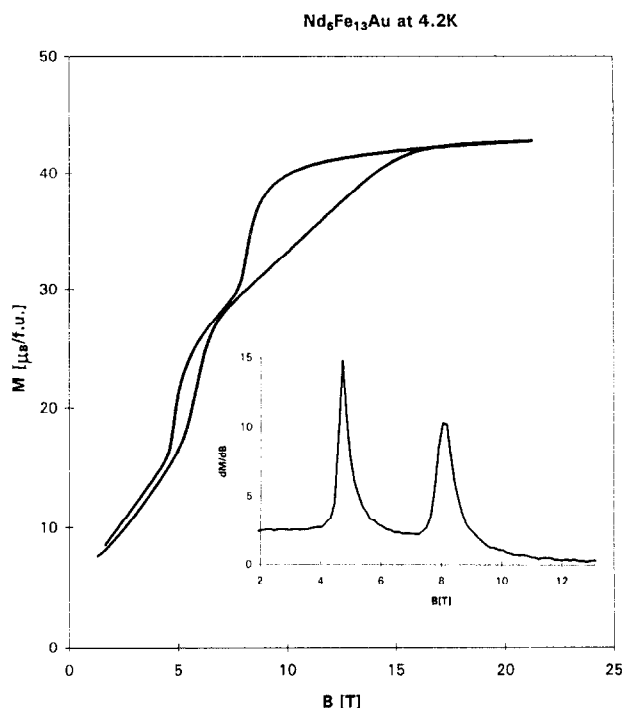
Fig. 2. Field dependence of the magnetic moment of $Nd_6Fe_{13}Au$ measured at 4.2 K with increasing and decreasing field.

Nd$_6$Fe$_{13}$Au 0.1Tesla

Fig. 1. Temperature dependence of the magnetisation of $Nd_6Fe_{13}Au$ measured in a field of 0.1 T.

the magnetic isotherm to zero field leads to a fairly small value of the magnetisation, indicating again that the magnetic structure of this compound consists of a number of magnetic sublattices with a mutually compensating contribution to the total magnetisation. Previous high-field studies made on isotypic compounds of the type $Nd_{6-x}La_xFe_{12}Ga_2$ have shown that the most likely magnetic structure at low temperatures is one in which the Nd sublattice orders antiferromagnetically and the Fe sublattice also orders antiferromagnetically [6]. It is apparent from the results shown in Fig. 2 that the prevailing antiparallel coupling is broken in two steps, the values of the corresponding critical fields being $B_{cr,1} = 4.7$ T and $B_{cr,2} = 8.2$ T.

When the high-field part of the magnetic isotherm of $Nd_6Fe_{13}Au$ is interpreted as corresponding to full ferromagnetic alignment of all participating magnetic moments, one can use the high-field moment in conjunction with the free-ion value for the Nd moments, to calculate the average value of the moment per Fe atom. This value is equal to 1.9 μ_B per Fe atom.

Mössbauer spectra obtained for $Nd_6Fe_{13}Au$ at various temperatures are shown in Fig. 3. It is seen that the Zeeman splitting of the spectra gradually decreases with increasing temperature when approaching T_N. Of particular interest is the fact that there is no sudden decrease of the Zeeman splitting when passing the temperature (340 K) at which the magnetisation decreases strongly in Fig. 1. Another interesting fea-
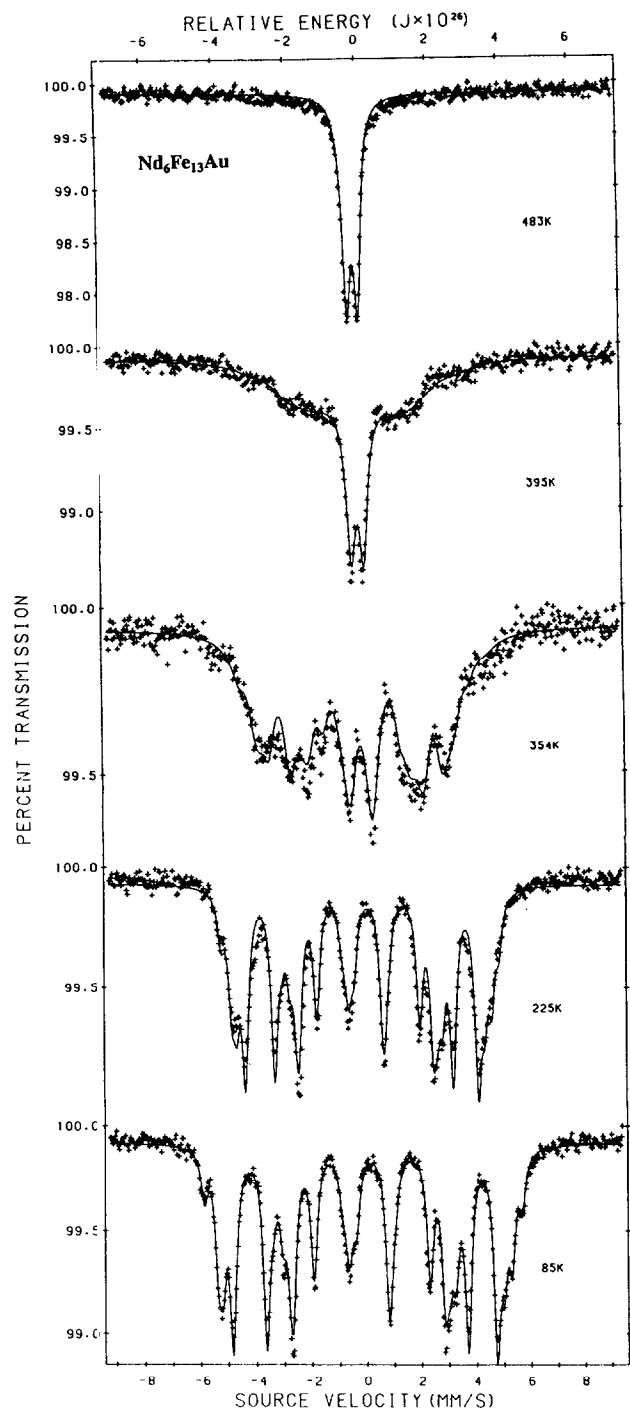
Fig. 3. Mössbauer spectra measured at various temperatures for $Nd_6Fe_{13}Au$.

Table 1
Mössbauer spectral results

TR (K)	δ^a_{ave} (mm s^{-1})	B_{ave} (T)	Magnetic area (%)
483	−0.19	0	0
435	−0.16	0	0
411	−0.16	2.7	17
395	−0.16	14.0	58
382	−0.15	15.8	50
369	−0.14	17.7	57
362	−0.14	18.2	59
354	−0.13	19.3	77
347	−0.12	20.6	93
340	−0.11	20.6	84
325	−0.10	20.8	96
295	−0.08	22.5	100
225	−0.03	25.5	100
155	−0.03	27.3	100
85	−0.05	28.8	100

^{a} Relative to room temperature α-iron foil.

possibilities is that concentration fluctuations lead to more gold-rich regions in which magnetic ordering occurs at a temperature lower than T_N.

The values of the average hyperfine field of the magnetically ordered components have been plotted as a function of temperature in Fig. 4. The shape of this curve indicates that magnetic ordering of the Fe sublattice sets in at T_N and proceeds in a normal way upon lowering the temperature. Only a minor discontinuity is observed around 340 K in the temperature dependence of the hyperfine field. This indicates that the strong rise found in the temperature dependence of the magnetisation around 340 K is not to be associated with a magnetic phase transition occurring
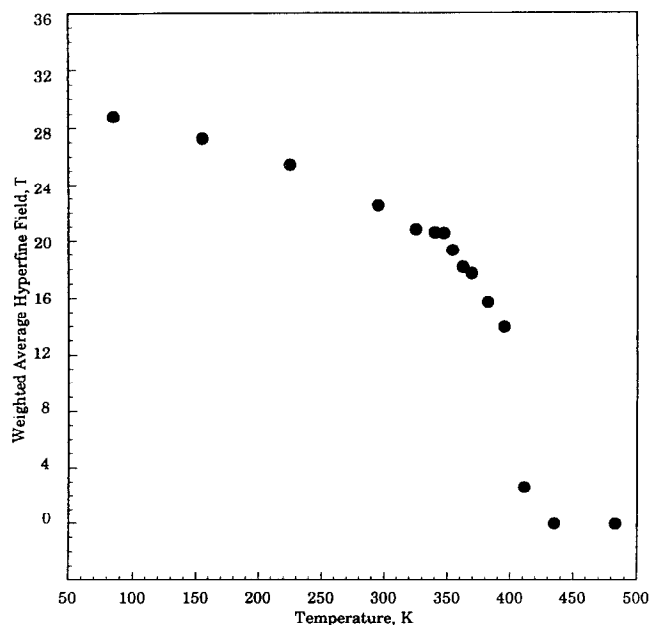
ture of the Mössbauer spectra is that a paramagnetic component develops with increasing temperature and strongly increases in relative absorption area as the temperature approaches T_N. The relative area of the ordered magnetic component, derived from fitting the Mössbauer spectra, is listed in Table 1, together with the average hyperfine field and the isomer shift. The exact origin of the paramagnetic component is still unclear and requires further investigation. One of the

Fig. 4. Temperature dependence of the average hyperfine field derived from fitting of the Mössbauer spectra of $Nd_6Fe_{13}Au$.

during the ordering process of the Fe moments in the compound $Nd_6Fe_{13}Au$.

From the high-field measurements we conclude that the fully ordered magnetic state of $Nd_6Fe_{13}Au$ is one in which the net magnetisation is very small. This means that the presence of very small amounts of a ferro- or ferrimagnetic impurity phase may manifest itself in a rather marked way in the overall temperature dependence of the magnetisation. We therefore believe that the strong rise of the magnetisation observed below approximately 340 K (see Fig. 1) is due to an impurity phase. As such the compound Nd_2Fe_{17} is the most likely candidate because its Curie temperature is about 330 K. In fact, it can be shown that the presence of about 1% of this phase is sufficient to cause the rise in magnetisation mentioned. Such small amounts can easily escape detection by both X-ray diffraction and Mössbauer spectroscopy. A similar interpretation of the temperature dependence of the magnetisation of $Nd_6Fe_{13}Cu$ was given earlier [7].

The average value of the hyperfine field derived from the Mössbauer spectra can be used to obtain an estimate of the magnitude of the average value of the moment per Fe atom in the compound $Nd_6Fe_{13}Au$. In an earlier investigation it was shown that a conversion ratio of 1.48 T/μ_B applies to a variety of different rare-earth based intermetallic compounds, including those with a fairly low Fe concentration [8]. Using this conversion ratio we find that the average Fe moment equals 1.9 μ_B at 85 K. This value is in good agreement with that derived from the high-field measurements at 4.2 K.

4. Conclusions

We have shown by means of magnetic measurements and Mössbauer spectroscopy that the magnetic ordering in $Nd_6Fe_{13}Au$ occurs at a much higher temperature than reported previously. Interestingly, the value of the average Fe moment of 1.9 μ_B found

from high-field measurements and Mössbauer spectroscopy in $Nd_6Fe_{13}Au$ is much higher than in the RFe_2 compounds, in which it is 1.5 μ_B per Fe atom, even though the Fe concentation is about the same. Of even more interest is the fact that the higher Fe moments lead to antiferromagnetic order ($T_N = 413$ K) in $Nd_6Fe_{13}Au$, as compared to ferromagnetic order ($T_C > 600$ K) in RFe_2. The occurrence of antiferromagnetic interactions in rare-earth based Fe compounds is of general interest and will be studied in more detail in future investigations.

Acknowledgements

References

[1] J. Allemand, A. Letand, J.M. Moreau, J.P. Nozières and R. Perrier de la Bathie, J. Less-Common Met., 166 (1990) 73.

[2] O.M. Sichevich, R.V. Lapunova, A.N. Soboley, Yu.N. Grin and Ya.P. Yarmoluk, Sov. Phys. Crystallogr., 30 (1985) 627.

[3] F. Weitzer, A. Leithe-Jasper, P. Rogl, K. Hiebl, A. Rainbacher, G. Wiesinger, J. Friedl and F.E. Wagner, J. Appl. Phys., 75 (1995) 7745.

[4] G.J. Long and F. Grandjean (eds.), in Supermagnets, Hard Magnetic Materials, Kluwer, Dordrecht, 1991, p. 355.

[5] R. Gersdorf, F.R. de Boer, J. Wolfrat, F.A. Muller and L.W. Roeland, in M. Date (ed.), High-field Magnetism, North Holland, Amsterdam, 1983, p. 277.

[6] Z.G. Zhao, C.H. de Groot, F.R. de Boer and K.H.J. Buschow, Physica B, 211 (1995) 102.

[7] T. Kajitani, K. Nagayama and T. Umeda, J. Magn. Magn. Mater., 117 (1992) 379.

[8] H.M. van Noort, D.B. de Mooij and K.H.J. Buschow, J. Appl. Phys., 57 (1985) 5414.