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Magnetic and Mössbauer spectral properties of the compound Nd₆Fe₁₃Au


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Abstract

The magnetic properties of Nd₆Fe₁₃Au 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 (Tₘ = 408 K). At low temperatures, the net moment of Nd₆Fe₁₃Au 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; Nd₆Fe₁₃Au; Magnetic properties; Mössbauer spectroscopy

1. Introduction

The compound Nd₆Fe₁₃Au crystallises in the tetragonal Nd₆Fe₁₂Si structure [1], which is an ordered variant of the La₆Co₁₁Ga₃ structure [2]. The magnetic properties of this compound were studied earlier by Weitzer et al. [3]. According to the latter authors the compound Nd₆Fe₁₃Au orders magnetically below Tₘ = 340 K, the magnetic ordering being accompanied by a strong rise of the magnetisation below Tₘ. 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 Nd₆Fe₁₃Au 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 Nd₆Fe₁₂Si 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
Nd₆Fe₁₃Au 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, reminiscent 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 \( \mu_B \) per formula unit of Nd₆Fe₁₃Au. 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 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 Ndₓ₋ₓLaₓFe₁₃Ga₂ 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 Nd₆Fe₁₃Au 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 \( \mu_B \) per Fe atom.

Mössbauer spectra obtained for Nd₆Fe₁₃Au 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-
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 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 around room temperature Fe-iron foil.

Table 1

<table>
<thead>
<tr>
<th>TR (K)</th>
<th>$\delta_{av}^s$ (mm s$^{-1}$)</th>
<th>$B_{av}$ (T)</th>
<th>Magnetic area (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>483</td>
<td>-0.19</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>435</td>
<td>-0.16</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>411</td>
<td>-0.16</td>
<td>2.7</td>
<td>17</td>
</tr>
<tr>
<td>385</td>
<td>-0.16</td>
<td>14.0</td>
<td>58</td>
</tr>
<tr>
<td>382</td>
<td>-0.15</td>
<td>15.8</td>
<td>50</td>
</tr>
<tr>
<td>360</td>
<td>-0.14</td>
<td>17.7</td>
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<td>362</td>
<td>-0.14</td>
<td>18.2</td>
<td>59</td>
</tr>
<tr>
<td>354</td>
<td>-0.13</td>
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<td>77</td>
</tr>
<tr>
<td>347</td>
<td>-0.12</td>
<td>20.6</td>
<td>93</td>
</tr>
<tr>
<td>340</td>
<td>-0.11</td>
<td>20.6</td>
<td>84</td>
</tr>
<tr>
<td>325</td>
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<td>96</td>
</tr>
<tr>
<td>295</td>
<td>-0.08</td>
<td>22.5</td>
<td>100</td>
</tr>
<tr>
<td>225</td>
<td>-0.03</td>
<td>25.5</td>
<td>100</td>
</tr>
<tr>
<td>155</td>
<td>-0.03</td>
<td>27.3</td>
<td>100</td>
</tr>
<tr>
<td>85</td>
<td>-0.05</td>
<td>28.8</td>
<td>100</td>
</tr>
</tbody>
</table>

$^a$ Relative to room temperature Fe-iron foil.

Fig. 3. Mössbauer spectra measured at various temperatures for Nd$_4$Fe$_{13}$Au.

Fig. 4. Temperature dependence of the average hyperfine field derived from fitting of the Mössbauer spectra of Nd$_4$Fe$_{13}$Au.
During the ordering process of the Fe moments in the compound Nd₈Fe₁₃Au.

From the high-field measurements we conclude that the fully ordered magnetic state of Nd₈Fe₁₃Au 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 Nd₈Fe₁₃Au 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 Nd₈Fe₁₃Cu 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 Nd₈Fe₁₃Au. In an earlier investigation it was shown that a conversion ratio of 1.48 T/μ₀ 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 μ₀ 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 Nd₈Fe₁₃Au occurs at a much higher temperature than reported previously. Interestingly, the value of the average Fe moment of 1.9 μ₀ found from high-field measurements and Mössbauer spectroscopy in Nd₈Fe₁₃Au is much higher than in the RFe₂ compounds, in which it is 1.5 μ₀ per Fe atom, even though the Fe concentration is about the same. Of even more interest is the fact that the higher Fe moments lead to antiferromagnetic order (Tᵣ = 413 K) in Nd₈Fe₁₃Au, as compared to ferromagnetic order (Tᵣ > 600 K) in RFe₂. 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.

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