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Investigation of the magnetic properties and $^{57}$Fe Mössbauer Effect in DyFe$_4$Ge$_2$

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Abstract

The magnetic properties of the compound DyFe$_4$Ge$_2$ have been studied by $^{57}$Fe Mössbauer spectroscopy and by magnetic measurements in high magnetic fields. The compound DyFe$_4$Ge$_2$ orders magnetically below 65 K. At 4.2 K it displays a strong field-induced first-order magnetic phase transition at $B_{c1} = 1.6$ T and a second weaker transition at $B_{c2} = 6.0$ T.

Keywords: DyFe$_4$Ge$_2$; Magnetic properties; Fe Mössbauer spectra

1. Introduction

Rare earth compounds of the composition RFe$_4$Ge$_2$ have been reported to crystallize in the tetragonal ZrFe$_4$Ge$_2$-type structure and to have interesting magnetic properties [1]. For DyFe$_4$Ge$_2$ a magnetic ordering temperature of 988 K has been observed, which is extremely high in view of the rather moderate Fe concentration of this compound. Attempts made by us to reproduce this high magnetic ordering temperature by measurements of the temperature dependence of the magnetization were unsuccessful. For this reason it was decided to study the magnetic properties of this material in more detail by means of high-field measurements and $^{57}$Fe Mössbauer spectroscopy.

2. Experimental

The sample of DyFe$_4$Ge$_2$ was prepared by arc melting from starting materials of at least 99.9% purity. The sample was wrapped in Ta foil and vacuum annealed in an evacuated quartz tube at 900 °C for four weeks. After this treatment the sample was investigated by X-ray diffraction. The latter measurements showed that the expected structure type [2] had formed and that the sample was approximately single phase, the impurity phase being of the ThCr$_2$Si$_2$ structure type and present in quantities of less than 5%.

The magnetic isotherms at 4.2 K were measured in the high-field installation at the University of Amsterdam [3] in fields up to 35 T.

3. Results and discussion

The $^{57}$Fe Mössbauer spectra were recorded on a constant acceleration type spectrometer with a $^{57}$Co–Rh source. For the calibration of the hyperfine fields we used $\alpha$-Fe$_2$O$_3$ at room temperature.

$^{57}$Fe Mössbauer spectra of DyFe$_4$Ge$_2$, recorded at room temperature and at 8 K, are shown in Fig. 1. It is immediately clear from the top spectrum that the compound is not magnetically ordered at room temperature. The low-temperature spectrum shows a substantial Zeeman splitting. It has been possible to fit this spectrum on the basis of two subspectra of equal intensity with slightly different hyperfine fields and different quadrupole splitting. The hyperfine parameters derived from the fitting procedure have been listed in Table 1. Because there is only a single crystallographic Fe site in the tetragonal ZrFe$_4$Ge$_2$ structure type the occurrence of the two subspectra is most conveniently interpreted as arising from an easy magnetization direction perpendicular to the c-axis and a concomitant magnetic splitting of the Fe sites.

$^{57}$Fe Mössbauer spectra of DyFe$_4$Ge$_2$ were obtained also at various other temperatures. The values of the hyperfine fields derived from fitting the spectra have been plotted as a function of temperature in Fig. 2. It follows from the results shown in this figure that the magnetic ordering temperature of DyFe$_4$Ge$_2$ is $65 \pm 1$ K.
Fig. 1. $^{57}$Fe Mössbauer spectra of DyFe$_4$Ge$_2$ at room temperature (top) and at 8 K (bottom).

Table 1

<table>
<thead>
<tr>
<th>Site 1</th>
<th>Site 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hyperfine field (T)</td>
<td>8.6</td>
</tr>
<tr>
<td>Quadrupole splitting (mm s$^{-1}$)</td>
<td>-0.025</td>
</tr>
<tr>
<td>Isomer shift (mm s$^{-1}$)</td>
<td>0.585</td>
</tr>
</tbody>
</table>

The average hyperfine field at 8 K is 7.9 T. If one assumes that the conversion factor 14.8 T/$\mu_B$ found earlier for various R–Fe compounds [4] does also apply to DyFe$_4$Ge$_2$ one may derive a value for the Fe moments in this compound equal to 0.53 \( \mu_B \).

The results of the high-field measurements at 4.2 K are displayed in Fig. 3. In the top part of the figure results are shown for a powder sample in which the powder particles were free to orient themselves into their equilibrium direction in the various field strengths applied. It is seen that at low field strength the moment is very small and tends to vanish in zero applied field. In the highest field strength applied the moment approaches the value 8 \( \mu_B \) per formula unit. As seen from the figure, the field-induced moment increase is not a continuous process but occurs by means of two discrete jumps at 1.6 T and 6.0 T. The first jump, in particular, is very pronounced and spans a moment increase of about 4 \( \mu_B \).

The results shown in the bottom part of Fig. 3 were obtained by fixing the random orientation of the powder particles with solidified ethanol before the high-field measurements. The latter were performed with increasing and decreasing fields. Both transition are seen
to display a substantial hysteresis, indicative of first-order magnetic phase transitions.

If one assumes that the Dy moment is equal to the free ion value $gJ/\mu_B = 10 \mu_B$ and bearing in mind that the Fe moments equal $0.53 \mu_B$, one can explain the vanishing spontaneous moment only by assuming that both sublattice moments are internally compensated by means of antiferromagnetic moment arrangements. Application of magnetic fields will break the antiferromagnetic moment arrangements eventually, although the present experimental information available does not allow us to further specify the magnetic phase transitions involved. It is interesting to note that the value of about 8 $\mu_B$ per formula unit observed in the highest field would correspond to a ferrimagnetic moment arrangement of the Dy sublattice moment and the Fe sublattice moment.

Concluding, we have shown that DyFe$_4$Ge$_2$ is not magnetically ordered above room temperature and one may expect a similar behaviour for the other compounds of the RFe$_4$Ge$_2$ series. The low magnetic ordering temperature is probably associated with the equally low value of the Fe moments. The magnetic structure of DyFe$_4$Ge$_2$ in zero or low field is more complicated than expected on the basis of a ferrimagnetic coupling between collinear ferromagnetic Dy and Fe sublattices.

References