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Magnetic interactions in $R_2(Fe_{1-x}Ga_x)_{17}$ ($R = Dy, Y$) compounds

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Abstract

By using X-ray diffraction and magnetic measurements, the structural and magnetic properties of $R_2(Fe_{1-x}Ga_x)_{17}$ ($R = Dy, Y$) compounds have been studied. It is shown that Ga substitution for Fe leads to an increase in lattice constants and a decrease in the average iron magnetic moment $\mu_{Fe}$ in these compounds, while the concentration dependence of the Curie temperature $T_C$ has a maximum. The exchange interaction constants $J_{DyFe}$ and $J_{FeFe}$ in $R_2(Fe_{1-x}Ga_x)_{17}$ ($R = Dy, Y$) compounds have been derived by means of mean-field analysis of $T_C$ and the values of $J_{DyFe}$ are consistent, with those derived from a mean-field analysis of the high-field magnetization. The behaviour of $T_C$ can be understood in terms of the concentration dependence of the exchange interaction constants.

1. Introduction

Intermetallic compounds of the type $R_2Fe_{17}$ ($R =$ rare earth or Y) have attracted much attention lately because of their ability to interstitially dissolve large amounts of nitrogen and carbon. This enhances their magnetic properties considerably and makes them interesting materials for permanent magnets [1,2]. To understand the influence of these interstitial atoms on the structural and magnetic properties, it is first necessary to understand better the interactions which are present in the parent compounds.

In this work, we have performed a study on the $R_2Fe_{17}$ ($R =$ Dy, Y) compounds with the substitution of Ga for Fe in the 3d sublattice, and focused our attention on the influence of such a substitution on the structural and the magnetic properties such as the Curie temperature $T_C$, the saturation magnetization $\sigma_s$, and the average iron magnetic moment $\mu_{Fe}$. The $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds were studied because Y is nonmagnetic, so that we can obtain some useful information about the magnetic properties, especially the exchange interaction of the 3d sublattice, which can be used in investigation of the other magnetic interactions in the $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds. Finally, the exchange interaction constants $J_{DyFe}$ and $J_{FeFe}$ in $R_2(Fe_{1-x}Ga_x)_{17}$ ($R =$ Dy, Y) were obtained in a mean-field analysis of $T_C$, and were then compared with the interaction constants obtained in mean-field analysis of the high-field magnetization curves.

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2. Experimental

$R_2(Fe_{1-x}Ga_x)_{17}$ ($R = Dy, Y$) compounds with $x = 0, 0.06, 0.08, 0.12, 0.16, 0.20, 0.30$ and $0.40$ were prepared by arc melting. The buttons were vacuum-annealed at $1273$ K for $10$ h X-ray diffraction has been performed to study the Ga concentration dependence of the crystal structure and the lattice parameters. The temperature dependence of the magnetization was measured by means of a vibrating sample magnetometer. The values of $T_c$ were determined from $\sigma^2 - T$ curves, where $\sigma$ is the magnetization measured in a low field ($B = 0.05$ T). At $4.2$ K, the magnetization curves of the $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds were measured using an extracting-sample magnetometer, and the high-field magnetization curves of $Dy_2(Fe_{1-x}Ga_x)_{17}$ were measured in the high-field installation at the University of Amsterdam [3]. The magnetic isotherms of powder particles, which were free to be oriented in the magnetic field, were recorded at $4.2$ K in external fields up to $35$ T. The saturation magnetizations were derived by $\sigma - 1/B$ plots extrapolated to infinite field.

3. Results and discussion

The X-ray diffraction patterns show that all the $R_2(Fe_{1-x}Ga_x)_{17}$ ($R = Dy, Y$) compounds are single phase, except for a very small amount of $\alpha$-$Fe$ as the second phase. The compounds crystallize in the $Th_2Ni_{17}$-type structure for $x \leq 0.12$ and in the $Th_2Zn_{17}$-type structure for $x \geq 0.20$; both structure types exist in $R_2(Fe_{0.84}Ga_{0.16})_{17}$. The X-ray diffraction patterns of $Dy_2(Fe_{1-x}Ga_x)_{17}$ with $x = 0, 0.08, 0.16$ and $0.20$ are shown in Fig. 1. The lattice constants $a$ and $c$, and the unit cell volumes of the $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds are listed in Tables 1 and 2, respectively. The values of the lattice constants $a$ and $c$ are also shown in Fig. 2.

In order to compare the $c$ values of the hexagonal cells with the rhombohedral ones, we have multiplied the former by $3/2$. It can be seen that the values of $a$ and $c$ increase monotonically with increasing Ga content in both the $Y_2(Fe_{1-x}Ga_x)_{17}$ and the $Dy_2(Fe_{1-x}Ga_x)_{17}$ series. The increase could be associated with the bigger radius of the Ga atom compared with the Fe atom. Figs. 3(a) and (b) show the magnetization curves for $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds, respectively. The $\sigma$
values have been corrected for the contribution of α-Fe impurity phase to the magnetization, which could be deduced from high-temperature magnetization. It can been seen that the saturation magnetization $\sigma_s$ decreases with increasing $x$. The $\sigma_s$ values of the $R_2(Fe_{1-x}Ga_x)_{17}$ ($R = Dy, Y$) compounds are plotted in Fig. 4 and are also listed in Tables 1 and 2, respectively. For comparison, the concentration dependence of $\sigma_s$ of both $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds derived in a simple dilution model is also shown in Fig. 4, which illustrates that as the Ga content increases, the decrease of $\sigma_s$ in both $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds is faster than expected in a simple magnetic dilution model. This is very similar to what occurs in $R_2(Fe_{1-x}Al_{1-x})_{17}$ ($R = Sm, Er$) compounds [4,5]. Since $Y$ atoms have no magnetic moment, the average iron magnetic moment $\mu_{Fe}$ in $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds could be obtained from the values of $\sigma_s$, as listed in Table 1, also shown in Fig. 5. The value of the average Fe magnetic moment decreases with Ga concentration, which may be caused by the transfer of valence electrons of the Ga atoms to the 3d subband of the Fe atoms, as is the situation of Al substitution for Fe [16,17]. The Ga concentration dependence of $T_C$ in both $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds is shown in Fig. 6, also listed in Tables 1 and 2. In both series of compounds, $T_C$ increases first with Ga content for small $x$, goes through a maximum at $x = 0.2$, and then decreases with increasing $x$, as observed in similar types of compounds [5,6]. Such a behaviour of $T_C$ may have two origins. The increase when $x < 0.2$ may be associated with the increase in the exchange interaction which results from the lattice

![Fig. 3. (a) Magnetization curves of $Y_2(Fe_{1-x}Ga_x)_{17}$ at 4.2 K. (b) High-field magnetization of $Dy_2(Fe_{1-x}Ga_x)_{17}$ at 4.2 K.](image)

![Fig. 4. Saturation magnetization at 4.2 K of $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds as a function of $x$.](image)
expansion upon Ga substitution. The decrease when \( x > 0.2 \) may be associated with the decrease in the iron magnetic moment.

The exchange interaction constants \( J_{FeFe} \) and \( J_{DyFe} \) were calculated by means of a mean-field analysis of \( T_c \). In general, a two-sublattice model can be used for the rare earth – transition metal (T) intermetallic compounds. There are three exchange interactions which can be described by three constants: \( J_{RR} \), \( J_{RT} \) and \( J_{TT} \). They decrease in the

---

**Table 1**

<table>
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<tr>
<th>( x )</th>
<th>( a )</th>
<th>( c )</th>
<th>( V )</th>
<th>( \sigma_s )</th>
<th>( \mu_{Fe} )</th>
<th>( T_c )</th>
<th>( J_{FeFe}/k_B )</th>
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**Table 2**

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<th>( a )</th>
<th>( c )</th>
<th>( V )</th>
<th>( \sigma_s )</th>
<th>( T_c )</th>
<th>( J_{DyFe}/k_B )</th>
<th>( J_{DyFe}/k_B )</th>
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sequence \( J_{TT}, J_{RT} \) and \( J_{RR} \), where \( J_{RR} \) is very small and can be neglected. The exchange interaction constants \( J_{ii} (i = R, T) \) are related to the molecular-field coefficient \( n_{ij} \) by the following expressions:

\[
\begin{align*}
 n_{ii} &= 2J_{ii}Z_{ii}(g_i-1)^2/g_i^2\mu_B^2 N_i \quad (i = R, T), \\
 n_{RT} &= 2J_{RT}Z_{RT}(g_R-1)(g_T-1)/g_R g_T \mu_B^2 N_T,
\end{align*}
\]

where \( N_i \) is the number of \( i \) atoms in per unit volume, \( Z_{ij} \) is the number of \( j \) atoms nearest neighbour to \( i \) atom.

An analysis of the Curie temperatures \( T_C \) of the rare earth–transition metal compounds gives:

\[
T_C = \left( T_{RR} + T_{TT} + \left[ (T_{TT} - T_{RR})^2 + 4T_{RT}^2 \right]^{1/2} \right)/2
\]

\((i, j = R, T),\)

where \( T_{ii} \) \((i = R, T)\) and \( T_{RT} \) can be written as:

\[
T_{ii} = 2J_{ii}Z_{ii}G_i/3k_B \quad (i = R, T),
\]

\[
T_{RT} = 2J_{RT}Z_{RT}G_R G_T/3k_B,
\]

with \( G_i = (g_i-1)^2J_i/(J_i+1) \) \((i = R, T)\). For \( \text{R}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \)[8], \( g_T = 2, Z_{RR} = 4, Z_{TT} = 10(1-x), Z_{RT} = 19(1-x), \mu_R = g_R \mu_B \mu_B \text{ and } \mu_T = 2S_T \mu_B \). In \( \text{Y}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds, \( T_{YY} = T_{FeFe} = 0 \) and \( T_{RT} = T_{FeFe} \). According to formula (3a), taking \( J_T = S_T, J_{FeFe} \) can be derived from \( T_C \) and the iron magnetic moment \( \mu_{Fe} \), as listed in Table 1. It can be seen that as the Ga concentration increases, \( J_{FeFe} \) increases first with Ga content, goes through a maximum at \( x = 0.3 \), and then decreases, as shown in Fig. 7. The increase of \( J_{FeFe} \) for \( x < 0.3 \) may be due to two reasons: first, Ga atoms may preferentially substitute for Fe atoms at some sites where the Fe atoms have negative exchange interaction with other Fe atoms; and second, the substitution of Ga for Fe increases the volume of the unit cell, as mentioned above. This may strengthen the Fe–Fe exchange interaction. The decrease in \( J_{FeFe} \) for \( x > 0.3 \) may be due to the decrease in the Fe concentration.

Using the values of \( J_{FeFe} \) deduced from \( \text{Y}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds, \( g_{FeFe} = 4/3, J_{y} = 15/2, \) the Curie temperature \( T_C \) and the average magnetic moment \( \mu_{Fe} \) of \( \text{Y}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \), the exchange interaction coefficients \( J_{DyFe} \) in the \( \text{Dy}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds were also derived, as shown in Fig. 7. It can be seen that the values of \( J_{DyFe} \) are nearly independent of the Ga content in the \( \text{Dy}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds.

On the other hand, the molecular-field coefficients \( n_{DyFe} \) in \( \text{Dy}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) were also derived by a mean-field analysis of the high-field magnetization [8], \( n_{DyFe} = 84\mu_0 \) for \( x = 0.3 \) and \( n_{DyFe} = 85\mu_0 \) for \( x = 0.4 \). Using formula (3b), we obtain the values \( J_{DyFe}/k_B = 8.25 \) K for \( x = 0.3 \) and \( J_{DyFe}/k_B = 8.68 \) K for \( x = 0.4 \), as shown in Fig. 7 by the black circles, which are in quite good agreement with those obtained by means of the molecular field analysis of \( T_C \).

4. Conclusions

The substitution of Ga for Fe in \( \text{Dy}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds leads to an increase in the lattice constants and a decrease in the average iron magnetic moment \( \mu_{Fe} \), while the concentration dependence of the Curie temperature exhibits a maximum. The exchange interaction constant \( J_{FeFe} \) between the Fe
spins exhibits a maximum at $x = 0.3$, whereas $J_{\text{DyFe}}$ is almost independent of the Ga content.

Acknowledgements

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References


