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

Y.H. Gao [a], N. Tang [a], X.P. Zhong [a], J.L. Wang [a], W.Z. Li [a], W.D. Qin [a], F.M. Yang [a,*], D.M. Zhang [b], F.R. de Boer [c]

[a] *Magnetism Laboratory, Institute of Physics, Academia Sinica, P.O. Box 603, Beijing 100080, China*
[b] *Physics Department, Huazhong University of Science and Technology, Wuhan 430074, China*
[c] *Natuurkundig Laboratorium, University of Amsterdam, 1018 XE Amsterdam, The Netherlands*

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Abstract

By using X-ray diffraction and magnetic measurements, the structural and magnetic properties of $R_2(Fe_{1-x}Gax)_{17}$ (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 μ_{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 σ_s, and the average iron magnetic moment μ_{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.

* Corresponding author.

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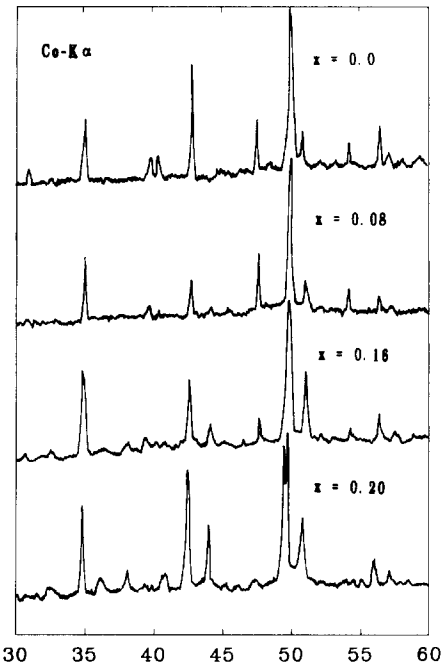

Fig. 1. X-ray diffraction patterns of $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds ($x = 0$, 0.08, 0.16 and 0.20).

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 σ 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 σ–$1/B$ 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 α-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 0.2 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 σ_s

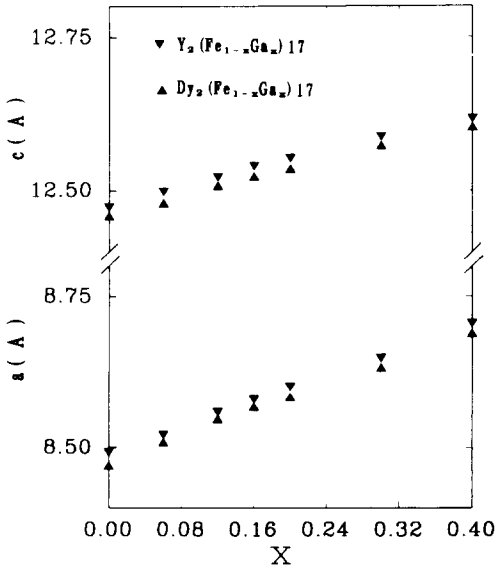

Fig. 2. Ga concentration dependence of the lattice constants a and c of $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds.

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 σ_s decreases with increasing x. The σ_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 σ_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 σ_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 $R_2(Fe_{1-x}Al_x)_{17}$ (R = Sm, Er) compounds [4,5]. Since Y atoms have no magnetic moment, the average iron magnetic moment μ_{Fe} in $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds could be obtained from the values of σ_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

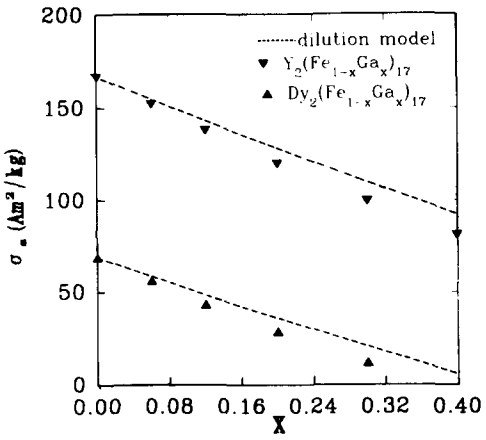

Fig. 4. Saturation magnetization at 4.2 K of $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds as a function of x.

$Dy_2(Fe_{1-x}Ga_x)_{17}$ 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

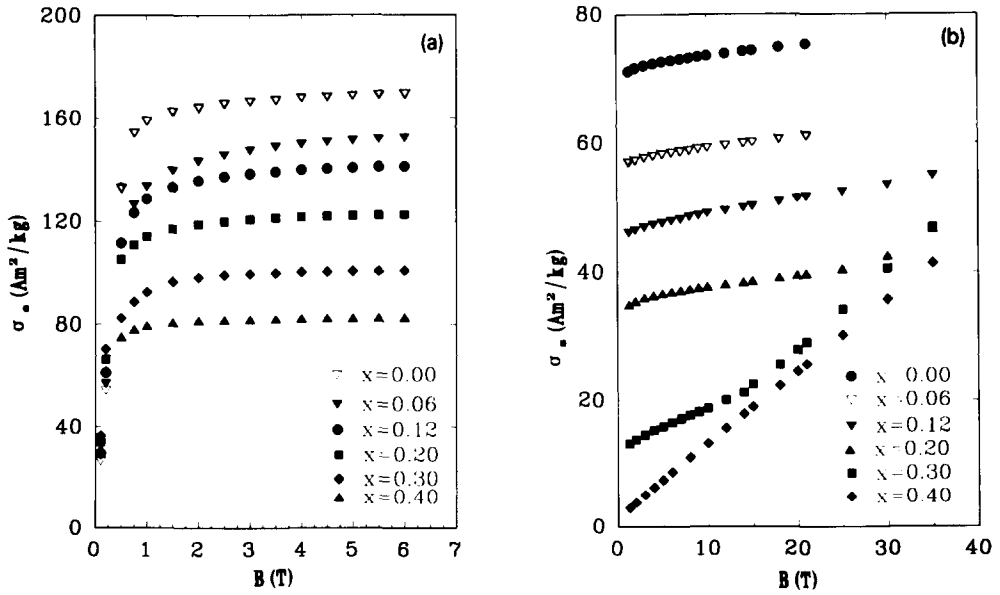

Fig. 3. (a) Magnetization curves of $Y_2(Fe_{1-x}Ga_x)_{17}$ at 4.2 K. (b) High-field magnetization of $Dy_2(Fe_{1-x}Ga_x)_{17}$ at 4.2 K.

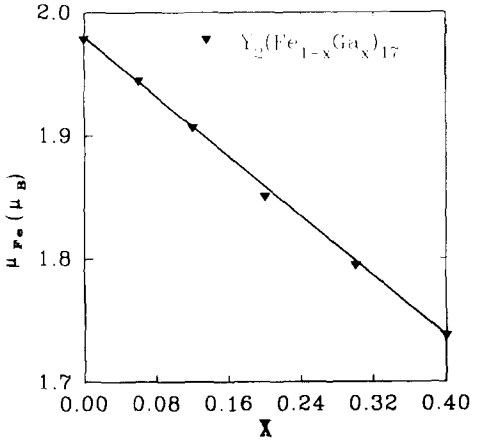

Fig. 5. Average Fe magnetic moment of $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds at 4.2 K as a function of x.

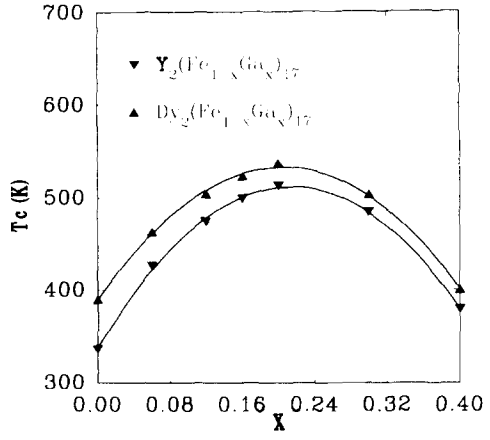

Fig. 6. Ga concentration dependence of the Curie temperature of $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds.

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
Lattice constants a and c, volume V, saturation magnetization σ_s, average iron magnetic moment μ_{Fe}, Curie temperature T_C, and Fe–Fe exchange interaction constant J_{FeFe} between Fe spins in $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds

x	a (Å)	c (Å)	V (Å^3)	σ_s (A m^2/kg)	μ_{Fe} (μ_B)	T_C (K)	J_{FeFe}/k_B (K)
0	8.492	8.315	519.28	173	2.05	336	24
0.06	8.521	8.332	523.90	157	2.01	426	34
0.12	8.559	8.348	529.60	142	1.96	475	42
0.16	8.580	8.360	532.97	–	–	499	–
0.20	8.601	12.553	804.20	123	1.90	513	52
0.30	8.650	12.588	815.66	103	1.86	484	58
0.40	8.706	12.618	828.22	83	1.78	379	56

Table 2
Lattice constants a and c, volume V, saturation magnetization σ_s, Curie temperature T_C, and exchange interaction constants (J_{1DyFe} from T_C, J_{2DyFe} from high-field magnetization) between Dy and Fe spins in $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds

x	a (Å)	c (Å)	V (Å^3)	σ_s (A m^2/kg)	T_C (K)	J_{1DyFe}/k_B (K)	J_{2DyFe}/k_B (K)
0	8.470	8.305	515.97	75.0	390	8.2	–
0.06	8.509	8.320	521.67	62.2	463	7.8	–
0.12	8.548	8.339	527.67	51.9	505	7.7	–
0.16	8.568	8.349	530.80	–	523	–	–
0.20	8.584	12.531	799.72	40.0	537	7.6	–
0.30	8.635	12.574	811.90	22.0	503	7.1	8.3
0.40	8.690	12.607	824.50	–	400	7.5	8.7

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:

$$n_{ii} = 2J_{ii}Z_{ii}(g_i - 1)^2/g_i^2\mu_B^2 N_i \quad (i = \mathrm{R, T}), \quad (1a)$$

$$n_{RT} = 2J_{RT}Z_{RT}(g_R - 1)(g_T - 1)/g_R g_T \mu_B^2 N_T, \quad (1b)$$

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:

$$T_C = \left\{T_{RR} + T_{TT} + \left[(T_{TT} - T_{RR})^2 + 4T_{RT}^2\right]^{1/2}\right\}/2 \quad (i, j = \mathrm{R, T}), \quad (2)$$

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

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

$$T_{RT} = 2J_{RT}\sqrt{Z_{RT}Z_{TR}G_R G_T}/3k_B, \quad (3b)$$

with $G_i = (g_i - 1)^2 J_i(J_i + 1)$ (i = R, T). For $R_2(Fe_{1-x}Ga_x)_{17}$ [8], $g_T = 2$, $Z_{RR} = 4$, $Z_{TT} = 10(1 - x)$, $Z_{RT} = 19(1 - x)$, $Z_{TR} = 2.5$, $\mu_R = g_R J_R \mu_B$ and $\mu_T = 2S_T\mu_B$. In $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds, $T_{YY} = T_{YFe} = 0$ and $T_C = T_{FeFe}$. According to formula (3a), taking $J_T = S_T$, J_{FeFe} can be derived from T_C and the iron magnetic moment μ_{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 $Y_2(Fe_{1-x}Ga_x)_{17}$ compounds, $g_{Dy} = 4/3$, $J_{Dy} = 15/2$, the Curie temperature T_C and the average magnetic moment μ_{Fe} of $Y_2(Fe_{1-x}Ga_x)_{17}$, the exchange interaction coefficients J_{DyFe} in the $Dy_2(Fe_{1-x}Ga_x)_{17}$ 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 $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds.

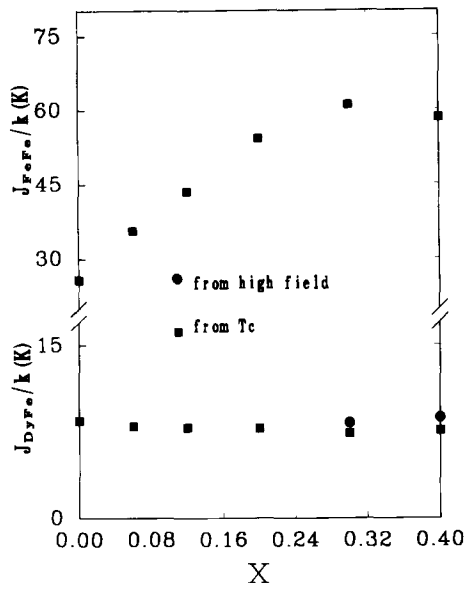

Fig. 7. Ga concentration dependence of the exchange interaction constants J_{FeFe} and J_{DyFe} in $Y_2(Fe_{1-x}Ga_x)_{17}$ and $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds.

On the other hand, the molecular-field coefficients n_{DyFe} in $Dy_2(Fe_{1-x}Ga_x)_{17}$ 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 3(b), 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 $Dy_2(Fe_{1-x}Ga_x)_{17}$ compounds leads to an increase in the lattice constants and a decrease in the average iron magnetic moment μ_{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_{DyFe} is almost independent of the Ga content.

Acknowledgements

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