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Magnetic properties of $R_2Fe_{17-x}Ga_x$ compounds ($R=Y, Ho$)

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Structural and magnetic properties of the $R_2Fe_{17-x}Ga_x$ compounds ($R=Y, Ho, 0 \leq x \leq 6$) have been investigated. All investigated $Ho_2Fe_{17-x}Ga_x$ compounds with $x > 2.5$ crystallize in the Th_2Zn_{17} -type structure and the others in the Th_2Ni_{17} -type structure. The substitution of Ga for Fe leads to an increase in lattice constants a , c , and unit-cell volume, a decrease of the average Fe moment, and a maximum of the Curie temperature as a function of the Ga concentration at $x=3$, for both $Y_2Fe_{17-x}Ga_x$ and $Ho_2Fe_{17-x}Ga_x$ compounds. The high magnetic field leads to a spin phase transition from the ferromagnetic to the canted phase. The critical field B_c of the transition decreases with increasing Ga concentration. The exchange interaction constants J_{TT} between Fe-Fe spins and J_{RT} between R-T spins have been derived from a mean field analysis of Curie temperature. It has been found that J_{TT} increases at first, going through a maximum at $x=3$, then decreases with increasing x , whereas J_{RT} is almost independent of the Ga content, which is consistent with the result obtained from a mean field analysis of the high field magnetization curves of the $Ho_2Fe_{17-x}Ga_x$.

I. INTRODUCTION

Since Coey and Sun reported that the introduction of nitrogen leads to a remarkable improvement in the magnetic properties of R_2Fe_{17} compounds,¹ which makes these compounds interesting materials for permanent magnets, the structural and magnetic properties of the interstitial nitrides have attracted much attention. In order to fully understand the interaction in these nitrides, it is necessary to first understand the exchange interaction of the R-T and T-T spins in their parent compounds. Previous studies of these interactions have shown improvement in the properties of these compounds by substituting other atoms for Fe or R.^{2,3} In this paper, the influence of the substitution of Ga for Fe on the magnetic properties of $R_2Fe_{17-x}Ga_x$ ($R=Ho, Y$) compounds has been studied. The exchange interaction of the R-T and T-T spins in these compounds has been calculated based on a mean field analysis of the Curie temperature of $R_2Fe_{17-x}Ga_x$ ($R=Y, Ho$), which are then compared with the results obtained by the mean field analysis of the high field magnetization curves of $Ho_2Fe_{17-x}Ga_x$.

II. EXPERIMENT

The $Ho_2Fe_{17-x}Ga_x$ ($x=0, 0.25, 1.0, 2.5, 4.0$, and 6.0) and $Y_2(Fe_{1-x}Ga_x)_{17}$ ($x=0, 0.06, 0.12, 0.20, 0.30$, and 0.40) compounds were prepared by arc melting the constituent elements of 99.9 wt % purity or better, and followed by annealing in an argon atmosphere at 1473 K for 4 h. X-ray diffraction was performed on powdered samples using $Cu-K\alpha$ radiation. Magnetization measurements at 4.2 K have been performed in the High Magnetic Field Installation at the University of Amsterdam.⁴ The measurements were done on powder particles about 30 μm , which are sufficiently small to regard them as monocrystalline. During the magnetization measurement, the particles are free to rotate in the sample holder, so that they can orient their magnetic moments into

the direction of the applied field. Magnetization versus temperature curves were measured in a field of 500 Oe by means of the vibrating sample magnetometer (VSM) between temperatures from 300 K to 800 K. The Curie temperature were derived by σ^2 vs T plot.

III. RESULT AND DISCUSSION

X-ray diffraction patterns showed that the $Ho_2Fe_{17-x}Ga_x$ compounds for $x \leq 2.5$ crystallize in Th_2Ni_{17} , and for $x > 2.5$ crystallize in Th_2Zn_{17} , as shown in Fig. 1. The substitution of a larger radius Ga atom for the Fe atom makes the Th_2Ni_{17} -type structure unstable. The values of lattice parameters a and c are given in Table I. The substitution of Ga for Fe results in a linear increase of the lattice constants in $Ho_2Fe_{17-x}Ga_x$ compounds, reflecting the larger radius of the Ga atom.

FIG. 1. The x-ray diffraction patterns of $Ho_2Fe_{17-x}Ga_x$; $x=0, 2.5, 4.0$, and 6.0 .

TABLE I. The lattice constants (a and c), Curie temperature (T_c), saturation magnetization M_s (Am^2/kg), and average iron magnetic moment (μ_{Fe}) for $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$ and $\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$ compounds. The data between parentheses a and c (± 0.005 Å); T_c (± 5 K); M_s (± 0.1 Am^2/kg); μ_{Fe} ($\pm 0.02 \mu_B/\text{Fe}$).

x	a (Å)	c (Å)	T_c (K)	M_s (Am^2/kg)	μ_{Fe} (μ_B/Fe)
Y_2Fe_{17}	8.492	8.315	337	166.6	1.98
$\text{Y}_2(\text{Fe}_{0.94}\text{Ga}_{0.06})_{17}$	8.522	8.332	426	152.0	1.94
$\text{Y}_2(\text{Fe}_{0.88}\text{Ga}_{0.12})_{17}$	8.559	8.348	475	137.8	1.91
$\text{Y}_2(\text{Fe}_{0.80}\text{Ga}_{0.20})_{17}$	8.601	8.369	513	119.7	1.85
$\text{Y}_2(\text{Fe}_{0.70}\text{Ga}_{0.30})_{17}$	8.650	8.392	484	99.5	1.79
$\text{Y}_2(\text{Fe}_{0.60}\text{Ga}_{0.40})_{17}$	8.706	8.412	379	81.0	1.74
$\text{Ho}_2\text{Fe}_{17}$	8.431	8.303	360	69.4	2.11
$\text{Ho}_2\text{Fe}_{16.75}\text{Ga}_{0.25}$	8.458	8.314	380	66.8	2.11
$\text{Ho}_2\text{Fe}_{16.0}\text{Ga}_{1.0}$	8.482	8.325	425	58.5	2.10
$\text{Ho}_2\text{Fe}_{14.5}\text{Ga}_{2.5}$	8.520	8.352	505	43.7	2.09
$\text{Ho}_2\text{Fe}_{13.0}\text{Ga}_{4.0}$	8.584	8.392	518	20.4	1.91
$\text{Ho}_2\text{Fe}_{11.0}\text{Ga}_{6.0}$	8.639	8.434	450	0.4	1.83

The Curie temperature T_c of the $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$ and $\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$ rises rapidly at first with x , going through a maximum at about $x=3$, and falls quickly with x , as shown in Fig. 2. The values of T_c for these compounds are also listed in Table I. The Curie temperature is mainly determined by the Fe–Fe exchange interaction, which is sensitive to the Fe–Fe distance. Therefore the increase of T_c for the compounds with $x < 2$ with x may be partially associated with the lattice expansion upon the substitution of Ga for Fe. On the other hand, it has previously been found that Ga, like Al in $\text{Y}_2\text{Fe}_{17-x}\text{Al}_x$ compounds, preferentially occupies the $6c$ sites.^{5–7} Therefore the substitution of Ga for Fe may decrease the negative exchange interaction between Fe–Fe at $6c$ sites and increase the Curie temperature of these compounds. The decrease of the Curie temperature for the compounds with $x > 3$ increasing with the Ga concentration is due to the decrease of the average iron magnetic moment and the reduced iron concentration.

The saturation moment of $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$ and $\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$ decreases with increasing Ga concentration. The average iron magnetic moment μ_{Fe} was derived on the basis of the saturation magnetization σ_s measured at 4.2 K. The σ_s values have been corrected for the contribution of

α -Fe impurity phase to the magnetization, which could be deduced from the high temperature magnetization measurements. In the evaluation of the magnetic moment per iron, it was assumed that the moment of Ho is independent of the Ga content, and the value is the same as that of the free Ho ion ($10\mu_B$). It can be found that the average Fe ion moment decreases with increasing Ga content, as shown in Table I.

In the rare earth-transition metal intermetallics, there are three types of the interactions, namely the R–R interactions between the magnetic moments within the R sublattice, the T–T interactions between the magnetic moments of T sublattice, and the R–T intersublattice interactions. Among them, the T–T interaction is the strongest and the R–R interaction is the weakest, and is usually neglected.⁸ According to the mean field analysis of Curie temperature,⁸ J_{RT} and J_{FeFe} can be expressed by

$$J_{\text{RT}} = g_{\text{R}} g_{\text{T}} \mu_B^2 N_{\text{T}} n_{\text{RT}} / 2 Z_{\text{RT}} (g_{\text{R}} - 1) (g_{\text{T}} - 1), \quad (1)$$

$$3kT_c = a_{\text{FeFe}} + (a_{\text{FeFe}}^2 + 4a_{\text{RFe}}a_{\text{FeR}})^{1/2}, \quad (2)$$

where

$$a_{\text{FeFe}} = Z_{\text{FeFe}} J_{\text{FeFe}} S_{\text{Fe}} (S_{\text{Fe}} + 1), \quad (3)$$

$$a_{\text{RFe}}a_{\text{FeR}} = Z_{\text{RFe}} Z_{\text{FeR}} S_{\text{Fe}} (S_{\text{Fe}} + 1) (g - 1)^2 J(J + 1) J_{\text{RFe}}^2, \quad (4)$$

$$J_{\text{FeFe}} = a_{\text{FeFe}} / Z_{\text{FeFe}} S_{\text{Fe}} (S_{\text{Fe}} + 1), \quad (5)$$

where a_{ij} is the microscopic exchange coupling parameters, J_{ij} is i – j exchange constants, n_{RT} expresses the macroscopic molecular field coefficients, $g_{\text{R}} = 8$ and $j = \frac{5}{4}$ for Ho, the orbital moment of the Fe atom is assumed to be quenched, so one takes $g_{\text{T}} = 2$, $J_{\text{T}} = S_{\text{T}}$, and $S_{\text{T}} = \mu_{\text{Fe}}/2$. Z_{ij} is the number of the nearest j neighbors of the i ion, N_i is the numbers of the i atom per formula unit. The J_{FeFe} and J_{RFe} were calculated using Eqs. (1)–(5). The values of J_{FeFe} and J_{RFe} as a function of Ga content are shown in Fig. 4. It can be seen that J_{RFe} is almost unchanged with x ; however, J_{TT} increases with the Ga content for $x < 3$, going through a maximum at about $x=3$, and then decreases for $x > 3$, as shown in Fig. 4. The J_{RT} can also be calculated from a mean field analysis of the high field magnetization curves.⁹ For ferrimagnetic R–T in-

FIG. 2. Ga content dependence of the Curie temperature T_c of $\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$ and $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$.

FIG. 3. The high field magnetization curves of $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$.

termetallics, in the fields beyond a critical field strength $B_{\text{cr},1} = n_{\text{RT}}[M_{\text{T}} - M_{\text{R}}]$ (M_{T} and M_{R} are the magnetization of the R and T sublattice, respectively), the exactly antiparallel R and T moments start to bend toward each other. The total magnetization is described by $M = B/n_{\text{RT}}$ and thus

$$\frac{dM}{dB} = [n_{\text{RT}}]^{-1}, \quad (6)$$

where $M = [M_{\text{R}}^2 + M_{\text{T}}^2 + 2M_{\text{R}}M_{\text{T}}\cos(\alpha)]^{1/2}$.

The parameter n_{RT} can be derived from the high field slopes for $B > B_{\text{cr},1}$. We can see in Fig. 3 that the critical field $B_{\text{cr},1}$ decreases with increasing Ga concentration because of the decrease of M_{T} . J_{RFe} was also calculated using the obtained values of n_{RT} and Eq. (1), and the values of J_{RT} are also shown in Fig. 4. They are in agreement with the results obtained by the mean field analysis of the Curie temperature.

IV. CONCLUSION

The substitution of Ga for Fe results in an increase in lattice constants and a transformation from the $\text{Th}_2\text{Ni}_{17}$ -type structure to the $\text{Th}_2\text{Zn}_{17}$ structure. A maximum of the Curie temperature with Ga concentration is due to the change of Fe-Fe and R-Fe exchange interaction and the decrease of average iron magnetic moment μ_{Fe} . The exchange interaction constants J_{TT} between Fe-Fe spins and J_{RT} between R-T spins have been derived, based on the mean field analysis

FIG. 4. Ga content dependence of the exchange interaction constants J_{FeFe} and J_{RFe} in $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$.

of Curie temperature. It has been found that J_{TT} increases at first, going through a maximum at $x=3$, then decreases with increasing x , whereas J_{RT} is almost independent of the Ga content, which is consistent with the result obtained from a mean field analysis of the high field magnetization curves of the $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$. J_{FeFe} is about four times J_{RFe} . The critical field $B_{\text{cr},1}$ decreases with increasing Ga concentration.

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