UvA-DARE (Digital Academic Repository)

A study on the exchange interaction in R₂Fe₁₇ compounds

Yang, F.M.; Wang, J.L.; Gao, Y.H.; Tang, N.; Han, X.F.; Pan, H.G.; Li, Q.A.; Hu, J.F.; Liu, J.P.; de Boer, F.R.

DOI

[10.1063/1.362399](https://doi.org/10.1063/1.362399)

Publication date

1996

Published in

Journal of Applied Physics

[Link to publication](#)

Citation for published version (APA):

Yang, F. M., Wang, J. L., Gao, Y. H., Tang, N., Han, X. F., Pan, H. G., Li, Q. A., Hu, J. F., Liu, J. P., & de Boer, F. R. (1996). A study on the exchange interaction in R₂Fe₁₇ compounds. *Journal of Applied Physics*, 79, 7883-7886. <https://doi.org/10.1063/1.362399>

General rights

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations

If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: <https://uba.uva.nl/en/contact>, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

A study on the exchange interaction in R_2Fe_{17} compounds

Fuming Yang, Jianli Wang, Yihua Gao, N. Tang, Xiufeng Han, Hongge Pan,
Li Qingan, and Jifan Hu

State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences, P. O. Box 603,
Beijing 100080, People's Republic of China

J. P. Liu and F. R. de Boer

Van der Waals-Zeeman Laboratory, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam,
The Netherlands

(Received 18 July 1995; accepted for publication 6 February 1996)

A study on the exchange interaction in R_2Fe_{17} compounds (R represents the heavy rare earth elements) has been performed by means of a mean-field analysis of the high-field magnetization curves, which were measured on the powder samples. Measurements have been carried out on R_2Fe_{17} -based quasiternary $R_2Fe_{17-x}M_x$ compounds with $R=Dy, Ho, \text{ and } Er$, $M=Al, Ga, \text{ and } Si$. It has been found that the value of the exchange coupling constant decreases with increasing atomic number of R ion of the compound. This behavior is explained by the varying $4f-5d$ hybridization in the compounds. © 1996 American Institute of Physics. [S0021-8979(96)10709-2]

I. INTRODUCTION

In the last three decades the rare earth (R) transition metal (T) intermetallic compounds have attracted great interest not only because of their significance for permanent-magnet application, but also due to the opportunity they offer for extensive fundamental studies of the magnetic behavior of $3d$ and $4f$ elements. Considerable attention has been given to the study on the magnetic interaction in these compounds. Three exchange interactions can be distinguished in the R-T compounds: the exchange interaction J_{TT} between the transition metal spins, J_{RT} between the rare earth spin and transition metal spins, and J_{RR} between rare earth spins. J_{TT} is dominant and responsible for the Curie temperature, which should be well above room temperature for application purposes. A strong R-T interaction is important to maintain the large magnetocrystalline anisotropy of the R ions up to the highest possible temperature and plays an important role in the formation of the magnetic structure. J_{RR} is the RKKY interaction and the strength is very weak compared with J_{TT} and J_{RT} , so that in the most cases it can be neglected.

In R-T intermetallic compounds the R-T exchange interaction is indirect consisting of the intra-atomic $4f-5d$ interaction and the interatomic $5d-3d$ interaction.¹ The exchange coupling constant $J_{ij}(i,j=R,T)$ can be investigated by different experimental methods, such as inelastic neutron scattering,² Mössbauer spectroscopy,³ first-order moment reorientation,⁴ molecular field analysis on the Curie temperature,⁵ and analysis of high-field magnetization process.⁶ Recently, following a model developed by Clark and Callen⁷ for ferrimagnetic garnets without magnetocrystalline anisotropy, Verhoef *et al.*⁸ have described the high-field magnetization behavior of the free powders (HFFP) of heavy-rare-earth transition-metal intermetallic compounds in very high magnetic field. By means of the HFFP method, values for J_{RT} can be derived straightforwardly from the high-field magnetization curves.

The aim of the present contribution is to investigate the R-T exchange interaction in R_2Fe_{17} -based compounds by means of the HFFP method.

II. EXPERIMENT

$R_2Fe_{17-x}M_x$ ($R=Dy, Ho, Er$, $M=Ga, Al, Si$, and $x=0, 1, 2, 3, \dots, 9$) alloys have been prepared by argon arc melting starting elements of purity at least 99.9%. Melting was performed in a water-cooled copper hearth and the ingots were remelted at least twice to promote homogeneity. Subsequently, the ingots were annealed in vacuum at 1473–1523 K for 4–8 h, wrapped in molybdenum foil and sealed in quartz tubes, followed by quenching to room temperature. X-ray diffraction with Co $K\alpha$ radiation was employed to check the phase and determine the lattice parameters of alloys. The Curie temperature of the investigated compounds were determined from σ^2-T plots by extrapolating σ^2 to zero. The magnetization curves $\sigma(B)$ were measured in an extracting-sample magnetometer in applied fields up to 7 T. The high-field magnetization was measured at 4.2 K in fields up to 35 in the High Field Installation at the University of Amsterdam.^{9,10} The measurements were performed on powder samples in which the powder particles were free to orient their magnetic moments parallel to the external field. The spontaneous magnetization values were derived by extrapolating the part of the magnetization curves corresponding to the antiparallel moment configuration to $B=0$.

III. RESULTS AND DISCUSSION

The x-ray diffraction patterns and the thermomagnetic analysis show that all samples investigated are single phase except for a small amount of α -Fe as second phase. For $x \leq 3$ all the compound crystallize in the Th_2Ni_{17} structure, for $x > 3$ in the Th_2Zn_{17} structure. The lattice parameters a and c were derived from the x-ray diffraction patterns. As an example, the structural parameters for the $Ho_2Fe_{17-x}Ga_x$ compounds are listed in Table I. The Curie temperatures T_C were

TABLE I. The lattice constants a and c , the Curie temperature T_C , the saturation magnetization σ_s , the average Fe atomic moment μ_{Fe} , the molecular field coefficient n_{RT} , and the exchange-couple constant of the compound $Ho_2Fe_{17-x}Ga_x$. The data in brackets were derived by extrapolating.

x	a (Å)	c (Å)	T_C (K)	σ_s (μ_B /f.u.)	μ_{Fe} (μ_B /Fe)	n_{RT} (10^{23} Tf.U/Am ²)	J_{RT}/k (K)
0	8.431	8.303	360	15.9	2.11	(2.48)	(6.9)
0.25	8.458	8.314	380	15.3	2.11	...	...
1.0	8.482	8.325	425	13.6	2.10	...	...
2.5	8.520	8.352	505	10.3	2.09	2.63	7.4
4.0	8.584	12.548	518	4.88	1.91	2.87	8.0
6.0	8.639	12.651	450	0.09	1.83	2.84	8.0
8.0	8.731	12.677	435	...	...	3.04	8.5

derived from the thermomagnetic curves. The values of T_C for the $Ho_2Fe_{17-x}Ga_x$ compounds are also listed in Table I.

The high field magnetization curves were measured at 4.2 K in fields up to 35 T. Figures 1 and 2 show the field dependence of the magnetic moment at 4.2 K of $Dy_2Fe_{17-x}Ga_x$ and $Ho_2Fe_{17-x}Ga_x$ free powder samples, respectively. It can be seen that the saturation magnetization obtained from the low field part of the magnetization curve decreases with increasing content of substituted atoms. This is due to the decrease of the moment of the Fe-sublattice resulting from the substitution of the nonmagnetic atoms. Since the Fe-sublattice moment is larger than the R-sublattice moment, the substitution of the nonmagnetic atoms leads to a decrease of the difference between the moments of the R and T sublattices. For small concentrations of substituted atoms, the magnetization remains approximately constant in the magnetic-field range used in the experiments. However, for larger concentrations a critical field $B_{cr,1}$ can be observed. The magnetization is approximately constant for B smaller than $B_{cr,1}$ but above $B_{cr,1}$ the magnetization increases nearly linearly with increasing applied field. The slope of this linear part and the values of $B_{cr,1}$ decrease with increasing content of substituted atoms.

In the case of free powders which can orient themselves into the minimum-free energy direction in the applied field,

the effect of the anisotropy on the magnetization process can be neglected if the anisotropy of at least one of the two sublattices can be neglected. In a molecular-field description, in that case the free energy of a ferrimagnetic compound with two magnetic sublattices R and T with magnetic moments M_R and M_T can be represented by

$$E_{RT} = n_{RT} M_R M_T \cos \alpha - (M_R + M_T) B. \tag{1}$$

The first term stands for the $3d-4f$ exchange interaction which contains the intersublattice-molecular-field coefficient n_{RT} and the angle α between the two sublattice magnetizations. The second term represents the magnetostatic energy (Zeeman energy) in the external field B . The equilibrium position of the magnetizations for each field strength can be found by minimizing the free energy. In the relatively low field condition, the moment configuration is strictly antiparallel and the magnetization has the value $M = |M_T - M_R|$. Beyond a critical field strength $B_{cr,1} = |M_T - M_R| n_{RT}$ the exactly antiparallel magnetic moments start to bend toward each other, and the total magnetic moment is described by

$$M = B / n_{RT} \tag{2}$$

with

$$M = (M_R^2 + M_T^2 + 2 M_R M_T \cos \alpha)^{1/2}. \tag{3}$$

FIG. 1. Field dependence of the magnetic moment of the $Dy_2Fe_{17-x}Ga_x$ compounds, measured on free powder at 4.2 K.

FIG. 2. Field dependence of the magnetic moment of the $Ho_2Fe_{17-x}Ga_x$ compounds, measured on free powder at 4.2 K.

FIG. 3. Concentration dependence of the exchange coupling constant $-J_{RT}/k$ in the $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$ compounds, as determined by HFFP. The solid line is a guide to the eye.

The value for n_{RT} can be thus derived from the slopes of straight line $M(B)$ obtained for $B > B_{cr,1}$. In even higher fields beyond a critical field strength $B_{cr,2} = |M_T + M_R|n_{RT}$, forced ferromagnetic alignment of the magnetic moments is achieved. The total magnetic moment then equals a value $M = M_R + M_T$. From the derived intersublattice-molecular-field coefficient n_{RT} , the related R-T exchange coupling constant J_{RT} in the Heisenberg Hamiltonian can be deduced,

$$J_{RT} = -N_T \mu_B^2 g_R n_{RT} / Z_{RT} (g_R - 1), \quad (4)$$

where Z_{RT} is the number of T neighbors surrounding the R atoms. N_T is the number of T atoms per formula unit. For $\text{R}_2\text{Fe}_{17-x}\text{Ga}_x$ compounds, $Z_{RT} = 19 - x$ and $N_T = 17 - x$. Using the value of n_{RT} obtained by Eq. (2) and the values of g_R of the rare earth, the value of J_{RT} can be derived for each compounds by Eq. (4). The derived values for n_{RT} and J_{RT} for the $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$ compounds are listed in Table I. The values of J_{RT} for the $\text{Ho}_2\text{Fe}_{17-x}\text{Ga}_x$ compounds are shown in Fig. 3 for several substitution contents. It can be seen that the value of J_{RT} shows a tendency to increase with increasing substitution content.

It is worth pointing out that the substitution of Al and Ga for Fe leads to a decrease of the average Fe atomic moment (see Table I). This may be associated with electron transfer

FIG. 4. The dependence of $-J_{RT}/k$ on the atomic number Z of R ions in the R_2Fe_{17} compounds, as determined by HFFP. The solid line is a guide to the eye.

from substitution atoms to the 3d band. By extrapolating the concentration dependence of J_{RT} to $x = 0$, the values of J_{RFe} for the nonsubstituted R-Fe intermetallic compounds were obtained. According to our results in the $\text{R}_2\text{Fe}_{17-x}\text{M}_x$ compounds the J_{RFe} derived by extrapolation to $x = 0$ for $M = \text{Ga}$ is almost equal to the values of J_{RFe} for $M = \text{Al}$ and Si . For example, the values of J_{RT} obtained from the $\text{R}_2\text{Fe}_{17-x}\text{Ga}_x$ compounds are listed in Table II for several heavy-rare-earth transition-metal intermetallic compounds. The values of J_{RT} obtained from magnetization measurements on single-crystalline R_2Fe_{17} compounds^{11,12} are also listed in Table II for comparison. Figure 4 shows the dependence of $-J_{RT}$ on the atomic number of the R component in the R_2Fe_{17} compounds. The values of $-J_{RT}$ obtained from magnetization measurements on single-crystalline R_2Fe_{17} compounds are also plotted in Fig. 4 for comparison. It can be seen that the values of J_{RFe} obtained by HFFP method are in agreement with those obtained by magnetization measurements on single-crystal samples. Both of them show the same tendency to decrease with increasing the atomic number Z of the R ion. This is associated with the lanthanide contraction. The decrease of the radius of 4f shell with increasing Z leads to a smaller overlap of the 4f and 5d shells, which results in a decrease of the 4f-5d hybridization and associated decrease of the R-Fe interaction J_{RFe} as a consequence.¹³

ACKNOWLEDGMENTS

The present investigation was supported by the National Nature Science Foundation of China and has been carried out within the scientific exchange program between China and the Netherlands.

TABLE II. The Curie temperature T_C , the spontaneous magnetization σ_s , the exchange-couple constant J_{RT}/k , derived from free powder and single-crystal samples of the R_2Fe_{17} compounds.

R	T_C (K)	σ_s (Am ² /kg)	J_{RT}/k (K)	
			Free powder	Single crystal
Gd	476	21.1	10.0	8.5 ^a
Tb	408	17.9	9.0	7.0 ^a
Dy	390	16.6	7.1	7.0 ^b
Ho	360	15.9	6.9	6.8 ^b
Er	313	14.5	6.2	6.4 ^b

^aReference 10.

^bReference 11.

¹I. A. Campbell, J. Phys. F Met. Phys. **2**, L47 (1972).

²K. N. Clausen and B. Lebech, J. Phys. C Cond. Matter **15**, 5095 (1982).

³P. C. M. Gubbens and K. H. J. Buschow, J. Phys. F Met. Phys. **12**, 2715 (1982).

⁴J. J. M. Franse, F. R. de Boer, P. H. Frings, R. Gersdorf, A. Menovsky, F. A. Muller, R. Radwanski, and S. Sinnema, Phys. Rev. B **31**, 4347 (1988).

⁵H. R. Kirchmayr and C. A. Poldy, in *Handbook on Physics and Chemistry*

- of *Rare Earth* edited by K. A. Jr. Gschneidner and J. Eyring (North-Holland, Amsterdam, 1979), Vol. 2, p. 55.
- ⁶R. Radwanski, J. J. M. Franse, and S. Sinnema, *J. Phys. F Met. Phys.* **15**, 969 (1985).
- ⁷A. E. Clark and E. Callen, *J. Appl. Phys.* **39**, 5972 (1968).
- ⁸R. Verstorff, R. Radwanski, and J. J. M. Franse, *J. Magn. Magn. Matter* **89**, 176 (1990).
- ⁹R. Gerstorff, F. R. de Boer, J. C. Wolfrat, F. A. Muller, and L. W. Poeland, *High-Field Magnetism*, edited by M. Date (North-Holland, Amsterdam, 1983), p. 277.
- ¹⁰R. Gersdorf, L. W. Roeland, and W. C. M. Mattens. *IEEE Trans. Magn.* **MAG-24**, 1052 (1988).
- ¹¹R. Verhoef, Ph.D. thesis, University of Amsterdam, 1990.
- ¹²S. Sinnema, Ph.D. thesis, University of Amsterdam, 1988.
- ¹³E. Bolorizky, M. E. Fremy, J. P. Gavigan, D. Givord, and H. S. Li, *J. Appl. Phys.* **61**, 3971 (1987).