Vol 13 No 11, November 2004
1009-1963/2004/13(11)/1965-04

Chinese Physics

© 2004 Chin. Phys. Soc.
and IOP Publishing Ltd

The anisotropy of Gd–Fe exchange interaction for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$*

Yan Yu(闫 羽)[a)b)], Xu Shu-Wei(许淑伟)[a)], Jin Han-Min(金汉民)[a)†], Du Xiao-Bo(杜晓波)[a)], and Su Feng(苏 峰)[a)]

[a)] *Department of Physics, Jilin University, Changchun* 130023, *China*

[b)] *State Key Laboratory for Superhard Materials, Jilin University, Changchun* 130023, *China*

(Received 7 April 2004; revised manuscript received 29 June 2004)

The magnetization curves along the crystal axes for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ were analysed based on the single-ion model. If the Gd–Fe exchange interaction has been taken as isotropic as usual, the fitted values of magneto-crystalline anisotropy of the Fe sublattices in Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ would become unreasonably different from those of the corresponding Y or Lu compounds. It was shown that the large difference is caused by the neglect of the anisotropy of the Gd–Fe exchange interaction.

Keywords: magnetization curves, magnetic anisotropy, rare-earth compounds
PACC: 7560E, 7530G, 7550B

1. Introduction

The magneto-crystalline anisotropy (MCA) of the Rn–T (Rn=nonmagnetic rare earths Y, La and Lu;T=Fe,Co) compound is the anisotropy of the T-sublattice in the R–T compound and differs little for different Rn.[1–7] Since Gd^{3+} ion is in an S state with negligibly small magneto-crystalline anisotropy, the MCA of a Gd–T compound is expected to be nearly the same as that of the corresponding Rn–T compound. Based on the single-ion model, we analysed the magnetization curves along the crystal axes measured at 4.2K on the single crystal of Gd_2Fe_{17} and its hydride $Gd_2Fe_{17}H_3$ using the values of Gd–Fe exchange interaction measured directly by inelastic neutron scattering (INS).[8] The calculations showed that the fitted values of MCA of the Fe sublattices in Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ are strikingly different from those of Lu_2Fe_{17} and $Lu_2Fe_{17}H_3$, respectively. A similar difference of anisotropy constants for Y_2Fe_{17} and Gd_2Fe_{17} is also observed.[9] Some authors of this paper found that the fitted values of MCA of the Co sublattices in the large MCA Gd–Co compounds, such as $GdCo_5$, Gd_2Co_7 and $Gd_2Co_{14}B$, are unreasonably different from those of the corresponding Rn–T compounds, and concluded that the large difference is ascribed to the anisotropy of the Gd–Co exchange interaction.[10,11] Gerard and Ballou [12] succeeded in explaining the magnetization curves of the $Gd(Co_{1-x}Ni_x)_5$ compounds by taking into account the anisotropy of the Gd–Co exchange interaction. In this connection, the Fe sublattices in R_2Fe_{17} and $R_2Fe_{17}H_3$ exhibit a large MCA, so that the MCA constant at 4.2K is 2.5K/(Fe atom) and 2.4K/(Fe atom) for Lu_2Fe_{17} and $Lu_2Fe_{17}H_3$,[7] respectively. Such a large MCA reveals that an appreciable part of the orbital angular moments of Fe atoms remains at some crystallographic sites. So it is expected that the R–Fe exchange interaction and the magnetic moments of Fe atoms in R_2Fe_{17} and $R_2Fe_{17}H_3$ are anisotropic. The apparent anisotropy of the magnetic moments of Fe atoms, indeed, has been observed in R_2Fe_{17} (R: Y, Lu and Gd)[6,7,13] and their hydrides $R_2Fe_{17}H_3$.[6,7]

This work will show that the anisotropy of the Gd–Fe exchange interaction in Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ is nonnegligible. If the anisotropy is neglected, the fitted values of the MCA constants for the Gd–Fe compounds would become unreasonably different from those of the corresponding Y or Lu com-

*Project supported by Specialized Research Fund for the Doctoral Programme of Higher Education of China and the National Natural Science Foundation of China (Grant No 10304006).

†E-mail: jhm@mail.jlu.edu.cn

pounds.

2. Method of analysis

Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ have a Th_2Ni_{17}-type hexagonal structure with space group $P6_3/mmc$. There are two sites for Gd ions. By neglecting the MCA of the Gd^{3+} ions and the Gd-Gd exchange interaction,[14,15] the Hamiltonian of the Gd ions averaged over the two crystallographic sites is simply the sum of Gd–Fe exchange interaction and Zeeman energy, i.e.

$$H = 2\mu_B \boldsymbol{H}_{ex} \bullet \boldsymbol{S} + 2\mu_B \boldsymbol{S} \bullet \boldsymbol{H}. \quad (1)$$

In the coordinate system with the x and z-axes along the a and c-axes, respectively, the Gd–Fe exchange interaction can be expressed as[10]

$$\boldsymbol{H}_{ex}(T) = n_{GdFe}\boldsymbol{M}_{Fe}(T)(1 + P\sin^2\theta_{Fe} + P'\sin^4\theta_{Fe}), \quad (2)$$

where n_{GdFe} is the molecular-field coefficient, $\boldsymbol{M}_{Fe}$ and θ_{Fe} are the magnetic moment of the Fe-sublattice per formula units (f.u.) and the angle between $\boldsymbol{M}_{Fe}$ and the c-axis, respectively; p and p' are the anisotropy constants of the Gd–Fe exchange interaction. $\boldsymbol{M}_{Fe}$ is anisotropic and is expressed as

$$\boldsymbol{M}_{Fe}(\theta_{Fe}, T) = \boldsymbol{M}_{Fe}(T)(1 - P_x(T)\cos^2\theta_{Fe}) \quad (3)$$

with P_x (4.2K)= 0.012 for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$.[6]

By diagonalizing the 8×8 matrix, the eigenstates and eigenvalues of the Hamiltonian, $|i\rangle$ and E_i (i=1,$\cdots$,8), are obtained for a given H and a given direction of $\boldsymbol{M}_{Fe}$. The free energy of Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ are given by

$$F(\boldsymbol{H}, \boldsymbol{M}_{Fe}, T) = -2kT\ln Z + K_1(T)\sin^2\theta_{Fe} + K_2(T)\sin^4\theta_{Fe} - \boldsymbol{M}_{Fe}(T) \bullet \boldsymbol{H}, \quad (4)$$

where

$$Z = \sum_i \exp(-E_i/kT). \quad (5)$$

Here, $K_1(T)$ and $K_2(T)$ are the MCA constants of the Fe-sublattice. $\boldsymbol{M}_{Fe}$ $(T/T_C)/\boldsymbol{M}_{Fe}(0K)$ and $K_1(T/T_C)/K_1(0K)$ for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ are assumed to be the same as those for Y_2Fe_{17} and $Y_2Fe_{17}H_3$ respectively.[6] The values of $\boldsymbol{M}_{Fe}$(4.2K), which are adjusted around the value of Lu_2Fe_{17} and $Lu_2Fe_{17}H_3$[7] for better fit, are taken to be 36.9 and 36.6μ_B/f.u. for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$, respectively. The equilibrium direction of $\boldsymbol{M}_{Fe}$ is determined from minimization of the free energy. The magnetic moments of the Gd ions and of the $Gd_2Fe_{17}H_x$ (x=0,3) are given by

$$\boldsymbol{M}_{Gd}(T) = -\mu_B \sum_i \langle i|\, 2\boldsymbol{S}\, |i\rangle \exp(-\boldsymbol{E}_i/kT)/Z, \quad (6)$$

$$\boldsymbol{M}(T) = \boldsymbol{M}_{Fe}(T) + 2\boldsymbol{M}_{Gd}(T). \quad (7)$$

The values of the Gd–Fe exchange interaction $2\mu_B H_{ex}$ (T=15K), which are measured directly from the inelastic neutron scattering experiment, are about 384 and 343K for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$, respectively.[8] Three sets of parameters p, p', K_1 (4.2K) and K_2 (4.2K) are used. In set 1 and set 2, the anisotropy of the Gd–Fe exchange interaction is neglected, i.e. $p = p' = 0$, and the values of K_1 (4.2K) and K_2 (4.2K) are obtained by fitting the calculations to the magnetization curves at 4.2K for set 1, and the values of the two parameters are those of Lu_2Fe_{17}[7] and $Lu_2Fe_{17}H_3$[7] for set 2. In set 3, the anisotropy of the exchange interaction is taken into account and the values of p and p' are obtained by fitting the magnetization curves at 4.2K with the values of K_1 (4.2K) for Lu_2Fe_{17} and $Lu_2Fe_{17}H_3$, respectively.

3. Results and discussion

Figures 1 and 2 show the magnetization curves of Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ along the c-axis and the basal plane at 4.2K, respectively. The symbols represent the experimental data of the single crystals, [6] and the solid and the dashed curves are the calculations for the parameters of set 1 and set 2, respectively. The maximum difference of the calculated magnetization $M(H)$ along c-axis by using the parameters of set 1 and set 3 is 0.01 μ_B/f.u. So the calculations for set 3 are essentially the same as those for set 1. It can be seen that the solid curves reproduce the experimental curves well, while the dashed curves do poorly.

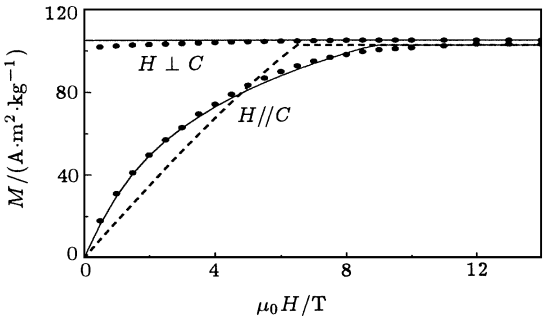


Fig.1. The magnetization curves of Gd_2Fe_{17} along the c-axis and along the basal plane at 4.2K: $\cdots$, experimental data taken from Ref.[6]; —,calculation by using the parameters of set 1; - - -, calculation by using the parameters of set 2.

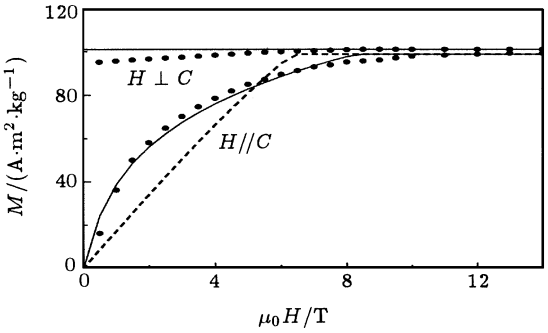


Fig.2. The magnetization curves of $Gd_2Fe_{17}H_3$ along the c-axis and along the basal plane at 4.2K: $\cdots$, experimental data taken from Ref.[6]; —, calculation by using the parameters of set 1; - - -, calculation by using the parameters of set 2.

Table 1. The values of $2\mu_B H_{ex}$, p, p', p_x, K_1, K_2 in Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ at T=4.2K.

Compound	set	$2\mu_B H_{ex}$/K	p	p'	p_x	K_1(K/f.u.)	K_2(K/f.u.)
Gd_2Fe_{17}	1	384.2	0	0	0.012	–62.0	19.0
	2	384.2	0	0	0.012	–42.0	0
	3	384.2	–0.0074	0.0071	0.012	–42.0	0
$Gd_2Fe_{17}H_3$	1	361.0	0	0	0.012	–57.0	22.0
	2	361.0	0	0	0.012	–40.0	0
	3	361.0	–0.0067	0.0087	0.012	–40.0	0

The three sets of the parameters for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ at T=4.2K are listed in Table 1. It can be seen from the table that the fitted MCA constants of the Fe-sublattice for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ (set 1) are apparently different from those for the corresponding Lu–Fe compounds (sets 2 and 3) if the anisotropy of the Gd–Fe exchange interaction is neglected. Table 1 also show that the fitted values of the anisotropy of the Gd–Fe exchange interaction in set 3, $4n_{GdFe}\mu_B \boldsymbol{SM}_{Fe}$ (4.2K) p and $4n_{GdFe}\mu_B \boldsymbol{SM}_{Fe}$ (4.2K) p', are just equal to the difference of K_1 (4.2K) and K_2 (4.2K) between set 3 and set 1 for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$. The relations can be deduced as follows. Since the calculated magnetization curves for set 3 are essentially the same as those for set 1, the free energies for the two sets should be the same. From Eq.(4) we have

$$\begin{aligned} F(\text{set } 1, 4.2\text{K}) \approx & 4n_{\mathrm{GdFe}}\mu_{\mathrm{B}}\boldsymbol{SM}_{\mathrm{Fe}}(4.2\text{K}) + 4\mu_{\mathrm{B}}\boldsymbol{S}\bullet H + K_1(\text{set } 1, 4.2\text{K})\sin^2\theta_{\mathrm{Fe}} \\ & + K_2(\text{set } 1, 4.2\text{K})\sin^4\theta_{\mathrm{Fe}} - \boldsymbol{M}_{\mathrm{Fe}}(4.2\text{K})\bullet\boldsymbol{H} \\ = F(\text{set } 3, 4.2\text{K}) \approx & 4n_{\mathrm{GdFe}}\mu_{\mathrm{B}}\boldsymbol{SM}_{\mathrm{Fe}}(4.2\text{K})(1 + p\sin^2\theta_{\mathrm{Fe}} + p'\sin^4\theta_{\mathrm{Fe}}) \\ & + 4n_{\mathrm{GdFe}}\mu_{\mathrm{B}}\boldsymbol{S}\bullet\boldsymbol{H} + K_1(\text{set } 3, 4.2\text{K})\sin^2\theta_{\mathrm{Fe}} \\ & + K_2(\text{set } 3, 4.2\text{K})\sin^4\theta_{\mathrm{Fe}} - \boldsymbol{M}_{\mathrm{Fe}}(4.2\text{K})\bullet\boldsymbol{H}. \end{aligned}$$

So we obtain

$$\begin{aligned} K_1(\text{set } 1, 4.2\text{K}) - K_1(\text{set } 3, 4.2\text{K}) &\approx 4n_{\mathrm{GdFe}}\mu_{\mathrm{B}}\boldsymbol{SM}_{\mathrm{Fe}}(4.2\text{K})p, \\ K_2(\text{set } 1, 4.2\text{K}) - K_2(\text{set } 3, 4.2\text{K}) &\approx 4n_{\mathrm{GdFe}}\mu_{\mathrm{B}}\boldsymbol{SM}_{\mathrm{Fe}}(4.2\text{K})p'. \end{aligned}$$

It is shown that the large difference between the fitted MCA constants of the Fe-sublattice for Gd_2Fe_{17} and $Gd_2Fe_{17}H_3$ and those of the corresponding Lu–Fe compounds is caused by the neglect of the anisotropy of the Gd–Fe exchange interaction.

References

[1] Alameda J M, Givord D, Lemaire R and Lu Q 1981 *J. Appl. Phys.* **52** 2079

[2] Bartashevich M I, Goto T, Yamaguchi M and Yamamoto 1995 *J. Magn. Magn. Mater.* **140/144** 855

[3] Abadia C, Algarabel P A, Garcia-Landa V, Ibarra M R, Moral A de, Kudrevatykh N V and Narkin P E 1998 *J. Phys. Condens. Matter* **10** 349

[4] Tereshina I S, Gaczynski P, Rusakov V S, Drulis H, Nikitin S A, Suski W, Tristan N V and Palewski T 2001 *J. Phys. Condens. Matter.* **13** 8161

[5] Tereshina I S, Nikitin S A, Pankratov N Yu, Bezkorovajnaya G A, Salamova A A, Verbetsky V N, Mydlarz T and Skourski Yu V 2001 *J. Magn. Magn. Mater.* **231** 213

[6] Tereshina I S, Nikitin S A, Skokov K P, Palewski T, Zubenko V V, Telegina I V, Verbetsky V N and Salamova A A 2003 *J. Alloys Compounds* **350** 264

[7] Tereshina I S, Nikitin S A, Stepien-Damm J, Gulay L D, Pankratov N Yu, Salamova A A, Verbetsky V N and Suski W 2001 *J. Alloys Compounds* **329** 31

[8] Isnard O, Sippel A, Loewenhaupt M and Bewley R 2001 *J. Phys.: Condens. Matter.* **13** 3533

[9] Takeda K, Maeda T and Katayama T 1998 *J. Alloys Compounds* **281** 50

[10] Jin H M and Yan Y 1993 *Phys. Rev.* B **48** 1022

[11] Yan Y, Jin H M and Wang X Q 1999 *Phys. Stat. Sol.* A **174** 505

[12] Gerard P and Ballou R 1992 *J. Magn. Magn. Mater.* **104/107** 1463

[13] Averbuch-Pouchot M T, Chevalier R, Deportes J, Kebe P and Lemair R 1987 *J. Magn. Magn. Mater.* **68** 190

[14] Zhang L G, Zhang S Y and Zhang H W 1997 *Acta Phys. Sin.* **46** 2241 (in Chinese)

[15] Shen B G, Liang B, Cheng Z H, Gong H Y, Zhang S Y and Zhang L G 1998 *J. Phys.* D **31** 2438