Hyperfine interactions in $R_x Gd_{1-x} Fe_3$ intermetallics R = Tb, Y

M. Kądziołka-Gaweł · A. Bajorek

Abstract The structural and magnetic properties of rare earth iron intermetallic compounds $Tb_x Gd_{1-x} Fe_3$ and $Y_x Gd_{1-x} Fe_3$ ($x = 0.0, 0.1, 0.2, 0.4, 0.5, 0.6, 0.8, 1.0$) was studied by X-ray diffraction, the ^{57}Fe Mössbauer effect and SQUID measurements. All investigated compounds crystallize in the rhombohedral PuNi3-type of crystal structure. The investigation of magnetic properties of $R_x Gd_{1-x} Fe_3$ proved their ferrimagnetic behavior. The Curie temperature of the investigated compounds decreases with the increase of R concentration from 721K ($GdFe_3$) to 655K ($TbFe_3$) and 533K (YFe_3). The saturation magnetic moment MS in the $R_x Gd_{1-x} Fe_3$ system increase with x parameter. The Mössbauer spectra are analyzed using four sextets, corresponding to three crystallographically (b, c, h) and four magnetically (b, c, h_1, h_2) inequivalent sites for iron. The mean hyperfine magnetic field increases with increase of the Gd concentration

Keywords Magnetic hyperfine interaction · Rare earth intermetallic compounds · Mössbauer studies

1 Introduction

The series of the RFe3 compounds where R is rare earth element exhibits very interesting magnetic properties due to the combination of 3d itinerant and 4f localized magnetism

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 Springer

[1–6]. Moreover, they attracted a great deal of attention not only by the academics but also due to their possible application as permanent magnet materials.

The magnetic properties in the RFe$_3$ series strongly depend on R atom. All these compounds except YFe$_3$ and SmFe$_3$ which are ferromagnets, indicate ferrimagnetic arrangement with a compensation point where the R and T magnetic moments equal zero or nearly zero [2]. In the whole series the resultant R and Fe moments are almost collinear. For the TmFe$_3$ compound the deviation from collinearity has been estimated as the largest. Analysis of the magnitude of f−d interactions based on the molecular field theory [2] indicates that the molecular field coefficients R−3d are stronger than R−R but weaker than the 3d−3d in all studied RFe$_3$ compounds except GdFe$_3$ and TbFe$_3$. Furthermore, the high Curie temperatures (T$_C$) confirm the dominant role of iron 3d exchange interactions.

In the ferrimagnetic GdFe$_3$ compound (T$_C$ = 729K) the existence of the compensation point between magnetic moments in Gd and Fe sublattices was evidenced at 618K. For the TbFe$_3$ compound T$_C$ is lower and equals 648K and for YFe$_3$ 569K [1, 7, 8].

As it was previously reported the values of the total magnetic moment varies with R, e.g in the case of GdFe$_3$, TbFe$_3$, DyFe$_3$, HoFe$_3$ and YFe$_3$ compounds equals $1.79\mu_B$, $3.13\mu_B$, $3.97\mu_B$, $4.53\mu_B$ and $5.24\mu_B$ respectively [1, 7–12]. Simultaneously the values of the iron moments varies with R, e.g in the case of GdFe$_3$, TbFe$_3$, DyFe$_3$, HoFe$_3$ and YFe$_3$ compounds equals $1.8\mu_B$, $1.79\mu_B$, $1.94\mu_B$, $1.73\mu_B$ and $1.67\mu_B$ respectively [1, 5–12]. The substitution within rare earth sublattice may lead to the changes of ff interactions which are mostly realized by 5d−3d hybridization.

By a complementary study of the magnetic properties for both series it is possible to explain the mechanisms responsible for the observed results. Thus, in the presented paper we are focused on influence of Gd/Tb as well as Gd/Y substitution on the magnetic properties of the R$_x$Gd$_{1-x}$Fe$_3$ compounds. In this paper the first detailed ^{57}Fe Mössbauer investigations on polycrystalline samples of Tb$_x$Gd$_{1-x}$Fe$_3$ and Y$_x$Gd$_{1-x}$Fe$_3$ ($x = 0.0, 0.1, 0.2, 0.4, 0.5, 0.6, 0.8, 1.0$) are presented.

2 Experimental details

The R$_x$Gd$_{1-x}$Fe$_3$ (R = Tb, Y) polycrystalline samples were prepared by arc-melting from high purity elements under argon atmosphere. The melted samples were then wrapped in tantalum foil, placed in quartz tubes and annealed at 900 °C for one week. The crystal structure of all samples was checked by means of X-ray diffraction (XRD) using Siemens D5000 diffractometer.

The magnetic properties of examined samples were measured with the use of SQUID magnetometer (MPMS XL7 Quantum Design). All measurements were performed in the 2K−400K temperature range and magnetic field up to 7T. In higher temperature range (300K−1050K) the DC measurements were performed by making use of the Faraday type magnetic balance in the magnetic field of 0.21T.

The MÖls sbauer absorption spectra were recorded at room temperature using a constant acceleration spectrometer with a ^{57}Co: Cr source of activity 50mCi. The metallic iron powder (α−Fe) absorber was used for velocity and isomer shift calibration of the Mössbauer spectrometer. These Mössbauer spectra were calculated by means of a discrete analysis (superposition of the Zeeman sextets).
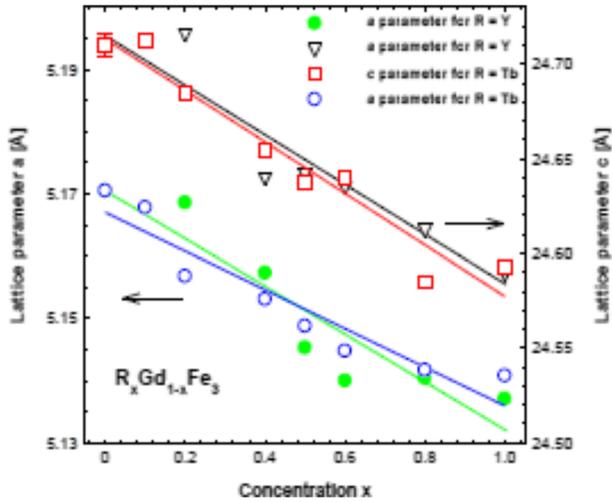
Fig. 1 The lattice constants versus x parameter for R concentration (R = Tb, Y) in the $R_x Gd_{1-x} Fe_3$ series

3 Results

The crystal structure of studied compounds was examined by making use of XRD method. Both series crystallize in the rhombohedral $PuNi_3$ type of crystal structure. This type of structure is presented in the literature as equivalent to hexagonal where a = b≠c and $\alpha = \beta = 90°$, $\gamma = 120°$ [1]. The rare earth atoms in the unit cell are located in two non-equivalent sites: R_I in 3a(0,0,0) and R_{II} in 6c (0,0,z) where z = 0.1414. The atoms of transition metals occupy three positions differ from each other by the lattice environment. They are located at T_I at 3b(0,0,1/2), T_{II} at 6c(0,0,z) where z = 1/3 and T_{III} at 18h (x,−x,z) where x = 1/2, x = 0.0829. The occupation of each position depends on the R content.

The crystal structure parameters of the $R_x Gd_{1-x} Fe_3$ (R = Tb, Y) series are presented in Fig. 1. We have observed a gradual decrease of the lattice parameters (a and c) as well as the volume of the unit cell with increasing of Tb/Y concentration. This behavior may be connected with different sizes of ionic radii for Gd (1.02 Å) and Tb (1.00 Å) and Y (0.93 Å). The obtained values of lattice parameters for the $GdFe_3$ and $TbFe_3$ compounds are in good agreement with those presented in the literature [1, 12].

Figure 2a presents the values of the Curie temperature for both $R_x Gd_{1-x} Fe_3$ series estimated from M(T) curves. Note, that the Curie temperature of the $R_x Gd_{1-x} Fe_3$ compounds decreases with the increase of R conce ntration from 721K ($GdFe_3$) to 655K ($TbFe_3$) and 533K (YFe_3). One can observe a much more faster decrease of T_C versus x content in the case of R = Y. It is an obvious behavior because yttrium can be considered as non-magnetic and does not carry a magnetic moment. So, if we substitute such an element in the rare earth matrix then the magnetic ordering can be much more weaker than we replaced Gd by the same amount of terbium, which indicates quite strong magnetism. It may suggest that the f−f exchange interaction becomes weaker in the case of yttrium dopant.
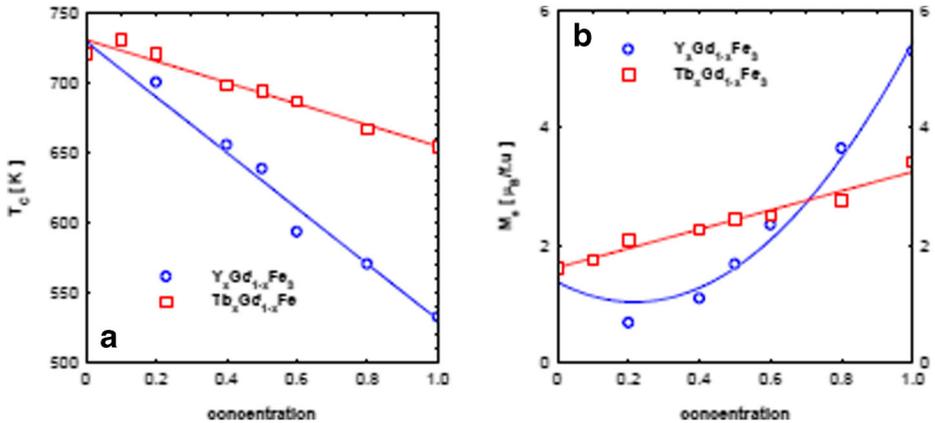
Fig. 2 Values of the (**a**) Curie temperature; **b** saturation magnetic moment of $R_x Gd_{1-x} Fe_3$ series as function of x parameter for R concentration (R = Tb, Y)

Figure 2b shows the values of the saturation magnetic moment $M_S(x)$ which has been estimated from the magnetic isotherms M(H) at 2K. The obtained results show that M_S varies almost linearly with the x parameter for terbium concentration. So, with Gd/Tb substitution the value of M_S gradually increases from $1.61\mu_B$/f.u ($GdFe_3$) to $3.42\mu_B$/f.u ($TbFe_3$). Also, the similar behaviour has been noticed for yttrium dopant. However, in this case such an increase is much more faster and M_S equals $5.32\mu_B$/f.u for YFe_3 compound. The observed $M_S(x)$ variation in both series may suggest the antiparallel type of coupling between R(4f) and Fe(3d) magnetic moments which strongly depend on the $4f - 3d$ exchange interactions and are realized via $4f-5d/4d$ as well as $5d/4d-3d$ interactions [5–8, 12]. So, the magnitude of the iron magnetic moment is related to the interaction within 3d band and between $4f-3d$ bands. In the $Tb_x Gd_{1-x} Fe_3$ system the f−d interaction and ff interaction seems to play an important role whereas in the $Y_x Gd_{1-x} Fe_3$ system with non-magnetic yttrium $3d-3d$ may largely determine the magnetic properties. It is worth to mention that in R = Y series a possible existence of ferromagnetic arrangement for a certain yttrium dopant (x≥0.2) may be observed. The values of the magnetic moment per Fe atom may be calculated from the values given for the saturation moment (at magnetic field 7T) after subtraction of the free gadolinium ($Gd^{3+} - 7\mu_B$) and terbium ($Tb^{3+} - 9\mu_B$) ions values and by assumption that yttrium is non-magnetic. So, the value of M_S/Fe atom obtained by such an estimation equals $1.80\mu_B$/Fe ($GdFe_3$), $1.86\mu_B$/Fe ($TbFe_3$) and $1.77\mu_B$/Fe (YFe_3).

It is worth to mention that in both cases such an estimation may be burdened by a certain error. This is due to the fact that in the series with R = Tb the value of terbium moment can be influenced by the crystal field (CEF) which have a significant influence on the magnetic properties. In the series with R = Y the yttrium atoms carry a small 4d magnetic moment.

Figure 3 shows selected room temperature Mössbauer spectra of the investigated compounds. Mössbauer spectra of $TbFe_3$, $GdFe_3$, YFe_3 are similar to those presented in literature [13]. As we know [13, 14], in magnetic RFe_3 metallic compound, the three crystallographically inequivalent iron sites (b, c, h) can give rise to four or more inequivalent magnetic positions of iron nuclei. It arises from, for example, to differences angles between the magnetic hyperfine field and the principal axes of the electric field gradient tensors. However, from Mössbauer measurements only, a determination of the easy axis in mixed crystal series seems to be very difficult. The Mössbauer absorption spectra of each investigated compounds are analyzed as a superposition of four Zeeman sextets (b, c, h_1, h_2)
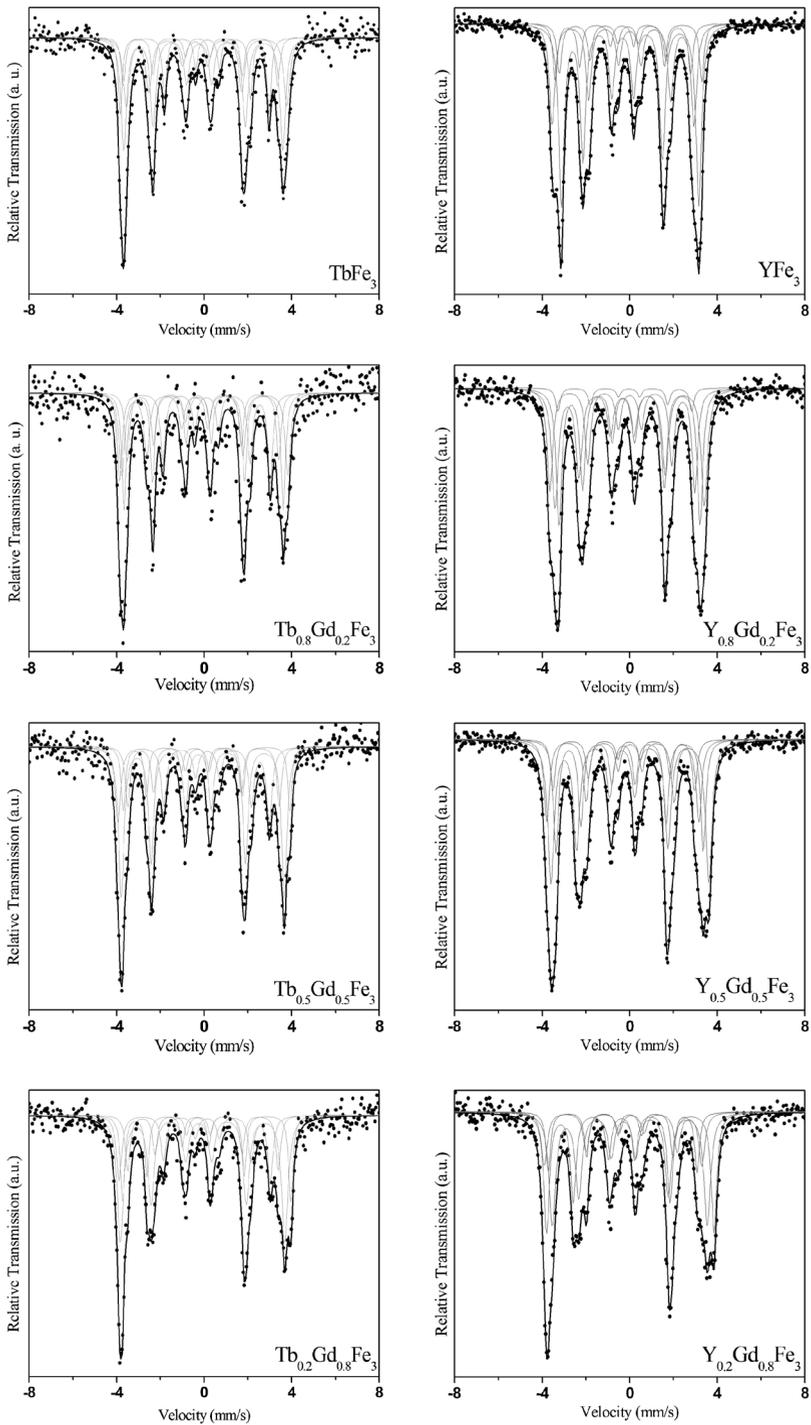
Fig. 3 Room temperature ^{57}Fe Mössbauer effect spectra of the selected $R_x Gd_{1-x}Fe_3$ (R = Tb,Y) intermetallics

Fig. 4 Mean magnetic hyperfine field B_{hf}, isomer shift IS and quadrupole splitting of $R_x Gd_{1-x} Fe_3$ series as function of x parameter for R concentration (R = Tb, Y)
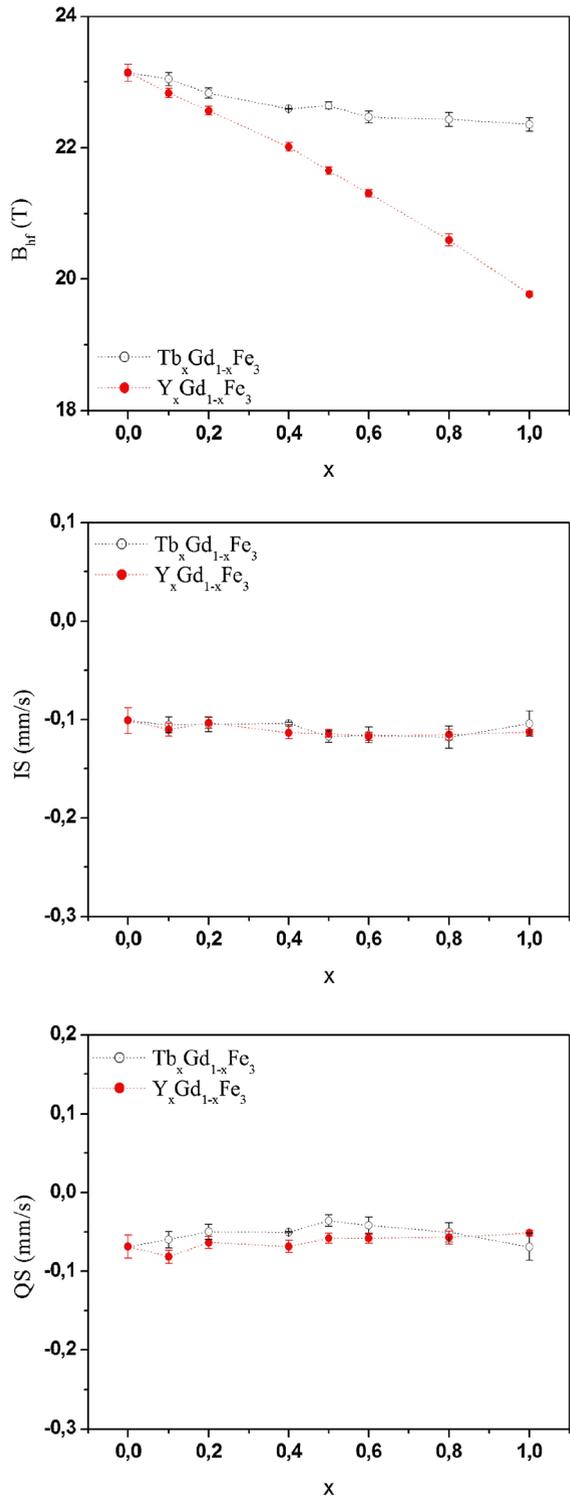
(Fig. 3). These spectra indicate on presence of long-range magnetic interaction at room temperature in investigated $R_xGd_{1-x}Fe_3$ (R = Tb, Y) series. Figure 4 contains the determined average hyperfine interaction parameters, i.e. the hyperfine magnetic field B_{hf} isomer shift IS and the quadrupole interaction parameter QS as function of x parameters of rare earth R concentration of $R_xGd_{1-x}Fe_3$ (R = Tb, Y). These parameters were determined as weighted average parameters of four components of each Mössbauer spectra. The values of the mean hyperfine magnetic fields determined for $Tb_xGd_{1-x}Fe_3$ are slightly higher then those obtained for $Y_xGd_{1-x}Fe_3$. The magnetic atom of the rare earth (Gd/Tb) leads to increase of ordering in Fe magnetic sublattice what cause increase of hyperfine magnetic fields on Fe atoms. The IS and QS values, in experimental error, change insignificant and do not depend on rare earth concentration.

4 Summary

Our magnetic and Mössbauer effect investigations confirm presence of magnetic long-range interaction at room temperature in $Y_xGd_{1-x}Fe_3$ and $Tb_xGd_{1-x}Fe_3$ (x = 0.0, 0.1, 0.2, 0.4, 0.5, 0.6, 0.8, 1.0) series. The results of magnetic measurements indicate on antiparallel tape of coupling between of rare earth sublattice and iron sublattice. However the increase of M_S value of higher yttrium dopant may suggest the presence of ferromagnetic arrangement. The Curie temperatures in the $R_xGd_{1-x}Fe_3$ compounds decrease linearly with increasing of concentration of R atoms in $GdFe_3$. The Mössbauer spectra of all investigated alloys are superposition of four Zeeman sextets what suggest four magnetically inequivalent position of Fe atom and indicate on that easy axis lie in the same direction in all $R_xGd_{1-x}Fe_3$ series.

References

1. Buschow, K.H.J.: Rep. Prog. Phys. **40**, 1179 (1977)
2. Herbst, J.F., Croat, J.J.: J. Appl. Phys **53**, 4304 (1982)
3. Burzo, E., Seitabla, D.: J. Phys. F: Metal Phys. **11**, L255 (1981)
4. Burzo, E., Seitabla, D.: J. Phys. F: Metal Phys. **12**, 2675 (1982)
5. Chrobak, A., Bajorek, A., Chekowska, G., Haneczok, G., Kwiecie, M.: Phys. Stat. Sol. (a) **206**, 731 (2009a)
6. Chrobak, A., Bajorek, A., Chekowska, G., Haneczok, G., Kwiecie, M.: J. Alloys. Compd. **485**, 6 (2009b)
7. Duc, N.H.: Intersublattice exchange coupling in the lanthanide-transition metal intermetallics. In: Gschneidner, K.A.Jr., Eyring, L. (eds.) Handbook on the Physics and Chemistry of Rare Earths. vol. 24, pp. 339–398. Elsevier (1997)
8. Duc, N.H., Hien, T.D., Givord, D., Franse, J.J.M., De Boer, F.R.: J. Magn. Magn. Matter. **124**, 305 (1993)
9. Tsai, S.C., Narasimhan, K.S.V.L., Kunesh, C.J., Butera, R.A.: J. Appl. Phys. **45**(8), 3582 (1974)
10. Narasimhan, K.S.V.L., Butera, R.A., Craig, R.S.: J. Appl. Phys. **44**(2), 879 (1973)
11. Simmons, M., Moreau, J.M., James, W.J.: J. Less Common Met. **30**, 75 (1973)
12. Bajorek, A., Chrobak, A., Ociepka, K., Chekowska, G.: Intermetallics **43**, 110–120 (2013)
13. Afril, S.K., Bunbury, D.St.P., Bowden, G.J., Day, R.K.: J. Phys. F: Metal Phys. **5**, 1048 (1975)
14. Gubbens, P.C.M., van der Kraan, A.M.: S. S. Commun. **26**, 107 (1978)