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Hyperfine fields at lanthanide nuclei in the intermetallic compounds of RFe_2

Y Li, C Carboni, J W Ross, M A H McCausland and D St P Bunbury
The Schuster Laboratory, The University, Manchester M13 9PL, UK

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Abstract. We have measured the hyperfine fields of ^{169}Tm in $\text{Tm}_{0.02}\text{Gd}_{0.98}\text{Fe}_2$ and $\text{Tm}_{0.02}\text{Y}_{0.98}\text{Fe}_2$ compounds using spin-echo NMR at 4.2 K. The results, together with complementary data obtained from other sources, are interpreted in terms of a simple model which involves hyperfine fields transferred from the lanthanide and iron ions. The effects of crystal-field quenching of the rare-earth moments in RFe_2 ($R = \text{heavy rare earth}$) and, where appropriate, magnetostriction, are estimated. We show that our simple model is satisfactory in the interpretation of hyperfine fields at lanthanide nuclei in the RFe_2 compounds. We obtain the coefficients $K_p = 9.4(1.4)$ T, $K_n = 3.5(0.9)$ T and $\beta = 15.8(1.6)$ T μ_B^{-1} .

1. Introduction

The rare-earth (R) intermetallic compounds RFe_2 , together with their pseudobinary variations, form a well-known isostructural series. These compounds have been extensively studied in the past few decades (see, for example, the review by Franse and Radwański 1993). Their simple structure (cubic Laves phase) makes them an ideal vehicle for studying the coexistence of 4f and 3d magnetism. Some of the intermetallic compounds are of technological importance, notably $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ (terfenol), which has good magnetostrictive properties.

In the light rare-earth compounds the R and Fe sublattices couple ferromagnetically; in the heavy rare earths the coupling is antiferromagnetic, leading to an overall ferrimagnetic structure. In the RFe_2 compounds the exchange interaction dominates the crystal-field interaction. However, the presence of crystal-field effects in the RFe_2 compounds is evident from many sources.

The RFe_2 compounds are well known for their giant magnetostriction (see, for example, Koon *et al* 1991). It is recognized that the magnetostriction of such compounds is highly anisotropic, exhibiting different magnetostrictive characters depending on the magnetization direction, i.e., $|\lambda_{111}| \gg |\lambda_{100}|$ (Clark 1979, 1980). The strict cubic symmetry of the R site may be broken by magnetostriction; under these circumstances the second-order crystalline potential arising from the magnetostrictive lattice strains should be considered (see, for example, Meyer *et al* 1979).

A number of NMR investigations have been devoted to the RFe_2 series and its pseudobinary compounds (Dormann 1991). First-principles calculations of the hyperfine fields at R nuclei in the RFe_2 compounds do not agree with the experimental values (see, for example, the work of Coehoorn and Buschow (1993) on GdFe_2). A satisfactory and conclusive picture of the hyperfine interactions for these compounds is not yet established.

In this paper we will use a simple phenomenological model which involves transferred hyperfine fields (THFF) arising from the R and Fe ions to describe the hyperfine fields at lanthanide sites. Our NMR results, together with the NMR and Mössbauer effect data obtained from other sources, are interpreted in terms of a theoretical framework which allows crystal-field quenching and, where appropriate, magnetostriction. We will show that our simple phenomenological model is satisfactory in the interpretation of hyperfine fields at lanthanide nuclei in the RFe₂ compounds.

In this paper we will only focus on the heavy rare-earth compounds RFe₂. Due to the fact that its valence electron configuration is similar to that of the lanthanides, YFe₂ is included.

2. Theoretical method

2.1. The electronic Hamiltonian

The interaction of the R ion with its environment may be expressed, within the mean-field approximation, by the effective electronic Hamiltonian

$$\mathcal{H}_{el} = \mathcal{H}_{cf} - \boldsymbol{\alpha} \cdot \mathbf{J} \quad (1)$$

where \mathcal{H}_{cf} represents the crystal-field interaction. In general, the vector $\boldsymbol{\alpha}$ includes contributions from the exchange, dipolar and applied fields (see, for example, McMorro *et al* 1989). In the system under consideration the dipolar field is negligible in comparison with the large exchange field arising mainly from the Fe sublattice; moreover, the measurements to be described were carried out in zero applied field, so we write

$$\boldsymbol{\alpha} = \boldsymbol{\alpha}_{ex} = -2\mu_B(g_J - 1)H_{exch} \quad (2)$$

where H_{exch} is the commonly used exchange field. The crystal-field interaction for R ions in cubic symmetry is expressed as follows (Hutchings 1964):

$$\mathcal{H}_{cf} = -\frac{2}{3}B_4^0(O_4^0 - 20\sqrt{2}O_4^3) + \frac{16}{9}B_6^0\left(O_6^0 + \frac{35}{4}\sqrt{2}O_6^3 + \frac{77}{8}O_6^6\right) \quad (3)$$

where B_4^0 and B_6^0 are the crystal-field parameters (CFP). It should be noted that in writing equation (3), it is assumed that the $\langle J_z \rangle$ coincides with the crystallographic [111] axis.

Magnetostriction can cause a distortion of the cubic symmetry at the R site. In the RFe₂ compounds the magnetostriction is highly anisotropic with $|\lambda_{111}| \gg |\lambda_{100}|$. The effect of magnetostriction is significant therefore only in those compounds in which the direction of easy magnetization is along [111] axis. Hence we may estimate K_{me} , the magnetoelastic contribution to the fourth-order anisotropy constant K_1 , as (Clark 1980)

$$K_{me} \simeq -\frac{9}{2}c_{44}\lambda_{111}^2 \quad (4)$$

where c_{44} is the elastic stiffness constant or modulus of elasticity, and λ_{111} the magnetostriction constant along [111] direction. Meyer *et al* (1979) have shown that magnetostriction can be taken into account by introducing a second-order term $B_2^0 O_2^0$ in the crystal-field Hamiltonian. Following Meyer *et al* (1979) B_2^0 may be estimated as

$$B_2^0 \simeq \frac{K_{me}}{3J(J - \frac{1}{2})}. \quad (5)$$

Hence in the presence of the magnetostriction effect the electronic Hamiltonian is written as

$$\mathcal{H}_{el} = -\alpha_{ex} J_z + B_2^0 O_2^0 - \frac{2}{3} B_4^0 (O_4^0 - 20\sqrt{2} O_4^3) + \frac{16}{9} B_6^0 \left(O_6^0 + \frac{35\sqrt{2}}{4} O_6^3 + \frac{77}{8} O_6^6 \right). \quad (6)$$

Provided that the exchange interaction dominates the crystal-field interaction, it is not necessary to diagonalize \mathcal{H}_{el} . We use a perturbation approach to estimate $\langle J_z \rangle$ in the presence of the $B_2^0 O_2^0$ -term as follows. \mathcal{H}_{el} in equation (6) may be partitioned into a sum of 'diagonal' and 'off-diagonal' terms

$$\mathcal{H}_{el} = \mathcal{H}_{diag} + \mathcal{H}' \quad (7)$$

where

$$\mathcal{H}_{diag} = -\alpha_{ex} J_z + B_2^0 O_2^0 - \frac{2}{3} B_4^0 O_4^0 + \frac{16}{9} B_6^0 O_6^0 \quad (8)$$

and

$$\mathcal{H}' = \frac{40\sqrt{2}}{3} B_4^0 O_4^3 + \frac{16}{9} B_6^0 \left(\frac{35\sqrt{2}}{4} O_6^3 + \frac{77}{8} O_6^6 \right). \quad (9)$$

A similar division has been used by Bleaney *et al* (1982). It is easily shown that the ground state of \mathcal{H}_{diag} is $|J_z = J\rangle$. As $\mathcal{H}_{diag} \gg \mathcal{H}'$, we employ perturbation theory to calculate the effect of \mathcal{H}' on \mathcal{H}_{diag} and obtain

$$\delta\langle J_z \rangle = J - \langle J_z \rangle = \sum_{M \neq J} \frac{J - M}{(E_J^0 - E_M^0)^2} |\langle M | \mathcal{H}' | J \rangle|^2. \quad (10)$$

The values of E_J^0 and E_M^0 are obtained from

$$\mathcal{H}_{diag} |J\rangle = E_J^0 |J\rangle \quad \mathcal{H}_{diag} |M\rangle = E_M^0 |M\rangle. \quad (11)$$

The formulae for calculating $\delta\langle J_z \rangle$ for $J = 6$ and $J = \frac{15}{2}$, which are used in this work, are given in the appendix.

2.2. The hyperfine interaction

In this work we discuss the hyperfine fields at lanthanide nuclei in the RFe₂ compounds. In the notation of McCausland and Mackenzie (1979), the hyperfine dipolar parameter a_t is a sum of intra- and extra-ionic contributions, respectively labelled with single and double primes:

$$a_t = a' + a'' \quad (12)$$

The dominant intra-ionic parameter is

$$a' = A \langle J_z \rangle \quad (13)$$

where A is the free-ion hyperfine coupling constant. The extra-ionic dipolar contribution a'' , which is small compared to a' , is related to the extra-ionic hyperfine field B'' by

$$a'' = \frac{\gamma}{2\pi} B'' \quad (14)$$

where γ is the gyromagnetic ratio of the R nucleus. Accordingly we may write the total hyperfine field B_t as

$$B_t = B' + B'' \quad (15)$$

where B' is given by

$$B' = \frac{2\pi}{\gamma} A \langle J_z \rangle. \quad (16)$$

In zero applied field the principal contributions to B'' may be written as

$$B'' = B_p + B_n^R + B_n^{Fe} \quad (17)$$

where B_p is the field due to conduction electrons polarized by the spin of the parent ion, B_n^R and B_n^{Fe} are commonly referred to as the THFF arising from conduction electrons polarized by the R and Fe neighbours, respectively. Following McCausland and Mackenzie (1979) we use a phenomenological model to describe the hyperfine fields. We assume that the conduction electron magnetization depends linearly on the projected spins of the parent ion and its neighbours. We also assume that B_n^{Fe} is proportional to the Fe moment. Hence equation (17) takes the form

$$B'' = K_p \langle \sigma_p \rangle + K_n \bar{\sigma} + \beta \mu_{Fe} \quad (18)$$

where $\langle \sigma_p \rangle = (g - 1) \langle J_z \rangle$ is the projected spin on the parent ion. $\bar{\sigma}$ denotes an average over all lanthanide spins in the host medium. K_p , K_n and β are constants for RFe₂ and its pseudobinary series.

3. Experimental details and results

The samples used in this work are of Tm_{0.02}Gd_{0.98}Fe₂ and Tm_{0.02}Y_{0.98}Fe₂. They were prepared by melting together the 99.99% pure starting materials in an argon-arc furnace. The ingots were annealed under argon for two weeks at 1000 °C, then quenched in oil. The cubic Laves structure was confirmed by x-ray diffraction. The material was powdered and embedded in epoxy resin prior to mounting in the coaxial resonator of a 2–8 GHz spectrometer (Carboni *et al* 1989). The spin-echo measurements were performed in zero applied field at 4.2 K.

The zero-field NMR spectra for Tm_{0.02}Gd_{0.98}Fe₂ and Tm_{0.02}Y_{0.98}Fe₂ are shown in figure 1. They each consist of single line, which is characteristic of ¹⁶⁹Tm ($I = \frac{1}{2}$). The resonance lines persist in fields up to 3 T. This suggests that the spectra obtained are attributable to nuclei within domains. The values of the hyperfine parameters a_t are given in table 1.

Table 1. Hyperfine parameters from the zero-field ¹⁶⁹Tm NMR spectra of Tm_{0.02}Gd_{0.98}Fe₂ and Tm_{0.02}Y_{0.98}Fe₂ at 4.2 K.

Compound	a_t (MHz)
Tm _{0.02} Gd _{0.98} Fe ₂	2466(2)
Tm _{0.02} Y _{0.98} Fe ₂	2422(2)

4. Discussion

The analysis of our NMR results, together with those obtained by other workers, may be outlined as follows. We first estimate the crystal-field quenching of the R moment in RFe₂ (R = Tb, Dy, Ho, Er, Tm) and its pseudobinary compounds. In the pseudobinary compounds of different probe R ions in the hosts GdFe₂ and YFe₂, we deduce the extra-ionic hyperfine

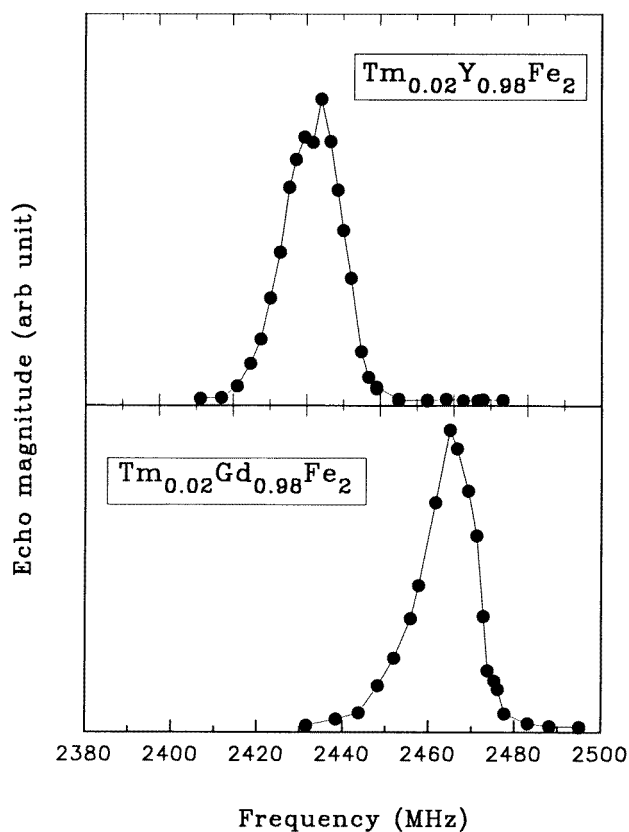


Figure 1. The zero-field NMR spectra of ^{169}Tm in $\text{Tm}_{0.02}\text{Y}_{0.98}\text{Fe}_2$ (top) and $\text{Tm}_{0.02}\text{Gd}_{0.98}\text{Fe}_2$ (bottom) at 4.2 K.

fields and use our phenomenological model of hyperfine fields to extract the coefficients K_p , K_n and β . Finally with the coefficients obtained we compare our calculated hyperfine fields with ones measured for $R\text{Fe}_2$ ($R = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}, \text{Tm}$).

The crystal-field parameters in the $R\text{Fe}_2$ compounds were derived from various experimental techniques, such as specific heat measurements (Germano and Butera 1981), neutron scattering studies (Koon and Rhyne 1981) and measurements of magnetocrystalline anisotropy (Atzmony and Dariel 1976). Unfortunately, for a given $R\text{Fe}_2$ compound there often exist different sets of crystal-field parameters and exchange fields H_{exch} obtained by different workers. For the purpose of this work we will give the average values of $\langle J_z \rangle$ calculated from different sets of crystal-field parameters and H_{exch} ; where appropriate, magnetostriction is included.

In table 2 the values of K_{me} are estimated from equation (4) where $c_{44} = 4.87 \times 10^{11}$ erg cm^{-3} (Rinaldi *et al* 1977). The variation of c_{44} is relatively small as a function of composition and temperature (Koon and Williams 1978). Note that K_{me} -values have been converted into units of K FU^{-1} (FU stands for formula unit) using the density data of Clark (1980). The values of B_2^0 are estimated from equation (5) for the compounds with their easy magnetization directions along the [111] axis. The values of $\langle J_z \rangle$ with and without considering the B_2^0 -term are given in table 2.

In table 2 we can see that the inclusion of magnetostriction (introducing the B_2^0 -term) has little effect on $\langle J_z \rangle$. We also notice that the quenching of the R moment is in most cases less than 0.3% ($\sim 1\%$ in TmFe_2). This result justifies the assumption made by many researchers that the rare-earth moment is fully polarized in the RFe_2 compounds at low temperatures (see, for example, Brooks *et al* (1991a) and Liu *et al* (1991)).

Table 2. The estimated quenching of the R moment in RFe_2 .

RFe_2	Easy direction (at 4.2 K)	$\lambda_{111}\dagger$ (10^{-3}) (at 0 K)	K_{me} (10^7) (K FU^{-1})	B_2^0 (K)	$\langle J_z \rangle$ (without B_2^0)	$\langle J_z \rangle$ (with B_2^0)
TbFe_2	[111]	4.4	-15.1	-0.15	5.9856(0.0067) ^a	5.9858(0.0068)
DyFe_2	[001]				7.4973(0.0002) ^b	
HoFe_2	[001]				7.9729(0.0136) ^a	
ErFe_2	[111]	-1.5	-1.7	-0.01	7.4954(0.0001) ^c	7.4957(0.0001)
TmFe_2	[111]	-3.7	-10.3	-0.10	5.9166(0.0242) ^d	5.9092(0.0299)

\dagger Taken from Clark (1980).

^a From Atzmony and Dariel (1976), Germano and Butera (1981) and Koon *et al* (1991).

^b From Atzmony and Dariel (1976) and Germano and Butera (1981).

^c From Atzmony and Dariel (1976), Koon and Rhyne (1978) and Germano and Butera (1981).

^d From Atzmony and Dariel (1976) and Bleaney *et al* (1982).

For future reference, we have listed in table 3 the saturation magnetization data for RFe_2 , μ_s , and the derived values of the Fe moment, μ_{Fe} . The magnetic moment of rare-earth ions per formula unit, μ_R , is obtained from $g\langle J_z \rangle$, where the values of g are after Bleaney (1988) (for intermediate coupling) and values of $\langle J_z \rangle$ are from table 2 with the B_2^0 -term considered where appropriate. Here we give the saturation magnetization data obtained from single crystals. The data for polycrystal GdFe_2 differ considerably, the quoted values ranging from 2.8 to 3.8 $\mu_B \text{FU}^{-1}$ (Buschow 1980). In table 3 we list 3.80 $\mu_B \text{FU}^{-1}$ for the magnetization value of GdFe_2 , the value chosen by Brooks *et al* (1991b) to achieve a general agreement between the calculated and measured magnetization data for the RFe_2 series. In table 3 we can see that the Fe moment is more or less constant at about 1.6 μ_B across the RFe_2 series.

The investigation of pseudobinary compounds such as $\text{R}_x\text{Y}_{1-x}\text{Fe}_2$ and $\text{R}_x\text{Gd}_{1-x}\text{Fe}_2$ is very useful. Here and afterwards we use the notation R:YFe_2 and R:GdFe_2 to denote that YFe_2 and GdFe_2 are hosts, and the R are probe ions. A typical formula is $\text{Tb}_{0.01}\text{Y}_{0.99}\text{Fe}_2$ or $\text{Tm}_{0.02}\text{Gd}_{0.98}\text{Fe}_2$.

So far as we are aware, no experimental data on magnetostriction in the pseudobinary compounds $\text{R}_x\text{Gd}_{1-x}\text{Fe}_2$ and $\text{R}_x\text{Y}_{1-x}\text{Fe}_2$ have been published. In $\text{Tb}_x\text{Gd}_{1-x}\text{Al}_2$ the magnetostriction coefficient λ_{111} is found to be proportional to x (Burd and Lee 1978). We assume that the above conclusion obtained for $\text{Tb}_x\text{Gd}_{1-x}\text{Al}_2$ may be also applicable to $\text{R}_x\text{Gd}_{1-x}\text{Fe}_2$ and $\text{R}_x\text{Y}_{1-x}\text{Fe}_2$ compounds. Hence in this way we may estimate the B_2^0 -term caused by magnetostriction to be of order 10^{-5} K in R:GdFe_2 and R:YFe_2 compounds, which is negligible.

There are no experimental data on measurements for exchange or crystal-field parameters for R:GdFe_2 and R:YFe_2 . But since the differences in lattice parameters are small, the crystal-field parameters for R ions in R:GdFe_2 and R:YFe_2 may be close to those for RFe_2 . We know that the exchange field on R ions arises mainly from the Fe sublattice, and that the Fe moment in RFe_2 is roughly constant. Hence the values of $\langle J_z \rangle$ for probe R moments in R:GdFe_2 and R:YFe_2 are estimated as the ones listed in column 6 of table 2.

In the following paragraphs we will discuss the hyperfine fields at lanthanide nuclei in R:GdFe_2 , R:YFe_2 and RFe_2 . As in section 2.2 the hyperfine intra-ionic field B' is

Table 3. Magnetization data for RFe₂.

RFe ₂	μ_s (μ_B FU ⁻¹)	μ_R (μ_B FU ⁻¹)	μ_{Fe} (μ_B /Fe)
GdFe ₂	3.80 ^a	6.97	1.59
TbFe ₂	5.82 ^b	8.94(0.01)	1.56(0.01)
DyFe ₂	6.87 ^b	9.92	1.53
HoFe ₂	6.69 ^b	9.90(0.02)	1.61(0.01)
ErFe ₂	5.79 ^b	8.96	1.59
TmFe ₂	3.72 ^b	6.88(0.03)	1.58(0.02)
YFe ₂	2.90 ^c	~0.4*	1.66*

^a Polycrystal data (Givord *et al* 1971). See the text.

^b Single-crystal data (after Clark 1980).

^c Polycrystal data (Buschow and van Stapele 1970, Givord *et al* 1971).

* Armitage *et al* (1986) and Dumelow *et al* (1986) have shown that $\mu_Y \simeq 0.4 \mu_B$, which is antiparallel to $\mu_{Fe} = 1.66 \mu_B$.

given by $B' = (2\pi/\gamma)A\langle J_z \rangle$ where the free-ion hyperfine coupling constants A are taken from Bleaney (1988) except $A = 528 \pm 3$ MHz for Tb³⁺ which is after Pelletier-Allard and Pelletier (1985). The values of γ are after Dormann (1991). The values of $\langle J_z \rangle$ are obtained from table 2.

For Gd³⁺ ions we take $B' = -32.2$ T (Bleaney 1988). The earlier treatment assumes that the Y hyperfine field purely arises from the conduction electron polarization (see, for example, Oppelt and Buschow 1973), which implies that there is no magnetic moment at the Y site and hence $B' = 0$. But Armitage *et al* (1986) and Dumelow *et al* (1986) have shown that in YFe₂ $\mu_Y \simeq 0.4 \mu_B$, which is antiparallel to $\mu_{Fe} = 1.66 \mu_B$. In YFe₂ $B' = -6.0$ T at the Y site is the estimate of Armitage *et al* (1989). Using the polarized neutron technique Ritter (1989) finds that $\mu_Y \sim 0.7 \mu_B$. In the following discussion we will analyse the available data using $\mu_Y \simeq 0.4 \mu_B$.

In table 4 we give the measured values of the hyperfine fields B_t , the estimated values of B' , the extra-ionic contribution B'' and $\langle \sigma_p \rangle$ in R:GdFe₂ and R:YFe₂ (R = Y, Tm, Ho, Tb, Gd). The calculated values of $\langle J_z \rangle$ are listed in table 2.

Table 4. Hyperfine fields (in T) and $\langle \sigma_p \rangle$ for R ions in R:GdFe₂ and R:YFe₂. The sign is defined with respect to the rare-earth moment.

R	B_t		B'	$B'' = B_t - B'$		$\langle \sigma_p \rangle$
	R:GdFe ₂	R:YFe ₂		R:GdFe ₂	R:YFe ₂	
Y	26.9(0.3) ^a	22.2(0.2) ^f	-6.0	32.9(0.3)	28.2(0.2)	0
Gd	42.4(0.2) ^b	30.6(0.5) ^g	-32.2	74.6(0.2)	62.8(0.5)	3.472
Tb	377.3(0.4) ^c	362.0(0.4) ^c	311.9(2.1)	65.4(2.1)	50.1(2.1)	2.951(0.003)
Ho	785.0(0.7) ^d	771.2(0.7) ^d	727.4(2.2)	57.6(2.3)	43.8(2.3)	1.929(0.003)
Tm	702.6(0.6) ^e	690.0(0.6) ^e	655.5(2.7)	47.1(2.8)	34.5(2.8)	0.970(0.004)

^a Vasil'kovskii *et al* (1974).

^b Extracted from the diagram of Vasil'kovskii *et al* (1988).

^c de Azevedo (1986).

^d Al-Assadi *et al* (1984).

^e This work.

^f Oppelt and Buschow (1976) and Riedi and Webber (1983).

^g Prakash *et al* (1983).

In the R:GdFe₂ series we notice that $\bar{\sigma} \simeq 3.5$ and $\mu_{Fe} \simeq 1.59 \mu_B$ (see table 3). Hence we may write equation (18) as

$$B'' = K_p \langle \sigma_p \rangle + (3.5K_n + 1.59\beta). \quad (19)$$

In the R:YFe₂ series we have $\bar{\sigma} \simeq 0$ and $\mu_{Fe} \simeq 1.66 \mu_B$ (see table 3); therefore we accordingly write equation (18) as

$$B'' = K_p \langle \sigma_p \rangle + 1.66\beta. \quad (20)$$

The value of $\langle \sigma_p \rangle$ depends on the probe ion R. From table 4 we plot out the variation of B'' with $\langle \sigma_p \rangle$ in figure 2 and derive the values of K_p , K_n , β in R:GdFe₂ and R:YFe₂. Best fits give $K_p = 8.8(1.2)$ T, $1.66\beta = 26.2(2.6)$ T in the R:YFe₂ series and $K_p = 9.9(0.8)$ T, $(3.5K_n + 1.59\beta) = 37.2(1.7)$ T in the R:GdFe₂. K_p , K_n and β are supposed to be constants through RFe₂, R:GdFe₂ and R:YFe₂; therefore we obtain

$$K_p = 9.4(1.4) \text{ T} \quad K_n = 3.5(0.9) \text{ T} \quad \beta = 15.8(1.6) \text{ T } \mu_B^{-1}. \quad (21)$$

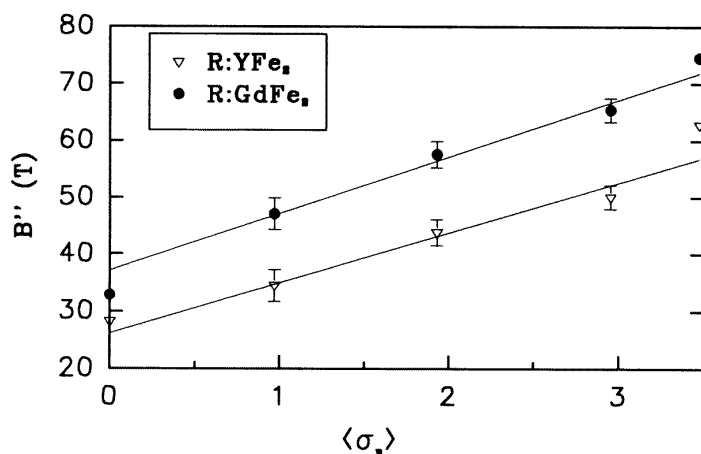


Figure 2. The variation of extra-ionic field B'' with the projected spin on the parent ion $\langle \sigma_p \rangle$ in R:GdFe₂ and R:YFe₂ (R = Y, Tm, Ho, Tb, Gd).

We are now in a position to calculate the hyperfine fields at lanthanide nuclei for the RFe₂ series (R = Gd, Tb, Dy, Ho, Er, Tm). As discussed before we have estimated the values of $\langle J_z \rangle$ for the RFe₂ compounds (see table 2); therefore the intra-ionic contribution B' can be obtained in a straightforward manner (see equation (16)). The extra-ionic contribution B'' in RFe₂ will be readily evaluated from equation (18) since the coefficients K_p , K_n and β have been already derived from R:GdFe₂ and R:YFe₂. Hence the total hyperfine fields B_t in RFe₂ can be evaluated, i.e., $B_t = B' + B''$. In table 5 we give the calculated values of B' , B_p , B_n^R and B_n^{Fe} , and the total hyperfine fields B_t^{calc} . We also list the experimental data on B_t for comparison.

In table 5 we have found that the calculated values of total hyperfine fields B_t^{calc} are in satisfactory agreement with the experimental ones B_t^{expt} except for for DyFe₂, where the uncertainty of the measured value is not given. In the following paragraph we will make use of the available NMR data to deduce the field contribution from the neighbouring lanthanide ions, B_n^R , and then make a comparison with the calculated values.

The predominant contribution of nearest neighbours (nn) to B_n^R has been observed in several intermetallic compounds (Dormann 1991). The satellite structures of ¹⁵⁷Gd NMR

Table 5. Calculated and measured hyperfine fields (in T) at lanthanide nuclei in the RFe₂ compounds. The sign is defined with respect to the rare-earth moment.

RFe ₂	B'	B_p^\dagger	$B_n^R^\dagger$	$B_n^{Fe^\dagger*}$	B_t^{calc}	B_t^{expt}
GdFe ₂	-32.2	32.6(4.9)	12.2(3.1)	25.1(2.5)	37.7(6.3)	42.4(1.0) ^a
TbFe ₂	311.9(2.1)	27.7(4.1)	10.3(2.7)	24.6(2.6)	374.5(5.9)	375.1(0.2) ^b
DyFe ₂	570.0(11.3)	22.8(3.4)	8.5(2.2)	24.2(2.4)	625.5(12.2)	648.0 ^c
HoFe ₂	727.4(2.2)	18.1(2.7)	6.8(1.7)	25.4(2.6)	777.7(4.7)	782.0(0.3) ^d
ErFe ₂	770.0(7.4)	13.8(2.1)	5.1(1.3)	25.1(2.5)	814.0(8.2)	818.4(1.0) ^e
TmFe ₂	654.7(3.3)	9.1(1.5)	3.4(0.9)	25.0(2.6)	692.2(4.6)	700.0(0.6) ^f

† These field contributions were calculated using the values of K_p , K_n and β ; see equations (17) and (18).

* $B_n^{Fe} = \beta\mu_{Fe}$ where the values of μ_{Fe} are from table 3.

^a Extracted from the diagram of Vasil'kovskii *et al* (1988) where the uncertainty is estimated by the authors. Gegenwarth *et al* (1967) give 45.3 T.

^b de Azevedo *et al* (1985).

^c Berthier *et al* (1981) where the uncertainty is not given.

^d Arif *et al* (1978).

^e Berthier and Devine (1981).

^f Carboni *et al* (1984) and Bleaney *et al* (1982).

in Gd_xY_{1-x}Fe₂ (Vasil'kovskii *et al* 1974) indicate that B_n^R has a pronouncedly short-range character. In GdPt₂ the four nn Gd atoms already contribute 80% of B_n^R (Dormann 1977). Here we assume that a roughly similar figure (~80%) may be applicable for the RFe₂ compounds. Using the experimental result that each nearest Gd neighbour produces a hyperfine field of 2.0 ± 0.2 T at the nucleus of another Gd ion (Vasil'kovskii *et al* 1974), we estimate that $B_n^{Gd} \approx 2.0(0.2) \times 4 \times \frac{100}{80} = 10.0(1.0)$ T in GdFe₂. Using the result that each nearest Er neighbour produces a hyperfine field of ~1.0 T at the nucleus of another Er ion from ¹⁶⁷Er NMR in Er_{0.9}La_{0.1}Fe₂ (Berthier and Devine 1981), we may accordingly estimate that $B_n^{Er} \approx 1.0 \times 4 \times \frac{100}{80} = 5.0$ in ErFe₂. Both of the estimated values agree well with our calculated B_n^R ; see table 5.

5. Conclusions

We have measured the hyperfine fields of Tm in Tm_{0.02}Gd_{0.98}Fe₂ and Tm_{0.02}Y_{0.98}Fe₂ compounds at 4.2 K. Using the available crystal-field parameters and allowing magnetostriction we have estimated the crystal-field quenching of R ions in the RFe₂ series. We conclude that the quenching of the R moment in most of RFe₂ is less than 0.3% (~1% in TmFe₂).

We have used a simple phenomenological model which involves hyperfine fields transferred from the lanthanide and iron ions and have derived the coefficients $K_p = 9.4(1.4)$ T, $K_n = 3.5(0.9)$ T and $\beta = 15.8(1.6)$ T μ_B^{-1} in the RFe₂ compounds. Using the estimated crystal-field quenching of the R moment and the coefficients obtained we have calculated the hyperfine fields at lanthanide nuclei in RFe₂. Agreement with the experimental values is good. Furthermore, we have the following findings: (I) the field contribution from the parent ions (B_p) is much larger than that from the neighbouring lanthanide ions (B_n^R); (II) the THFF from the Fe sublattice (B_n^{Fe}) is more or less ~25 T. In conclusion we have shown that our simple model is satisfactory in the explanation of hyperfine fields at lanthanide nuclei in RFe₂.

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Appendix

Using the matrix elements tabulated by Hutchings (1964) we derive the formulae for calculating $\delta\langle J_z \rangle$ (see equation (10)) by the perturbation method as follows:

$$\delta\langle J_z \rangle = J - \langle J_z \rangle = \frac{3A}{B} + \frac{6C}{D}$$

where for $J = 6$

$$\begin{aligned} A &= (360\sqrt{110}B_4^0 + 47\,040\sqrt{110}B_6^0)^2 \\ B &= (-3\alpha_{ex} + 81B_2^0 - 6120B_4^0 - 28\,224B_6^0)^2 \\ C &= (12\,320\sqrt{231}B_6^0)^2 \\ D &= (-6\alpha_{ex} + 108B_2^0 - 600B_4^0 + 833\,280B_6^0)^2 \end{aligned}$$

and where for $J = \frac{15}{2}$

$$\begin{aligned} A &= (240\sqrt{910}B_4^0 + 61\,600\sqrt{910}B_6^0)^2 \\ B &= (-3\alpha_{ex} + 108B_2^0 - 18\,960B_4^0 + 147\,840B_6^0)^2 \\ C &= (6160\sqrt{386.43}B_6^0)^2 \\ D &= (-6\alpha_{ex} + 162B_2^0 - 5760B_4^0 + 2217\,600B_6^0)^2. \end{aligned}$$

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