^{89}Y-NMR STUDY OF HYPERFINE INTERACTIONS IN $(R_x Y_{1-x})Fe_2$[**]

by

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[89]Y pulsed NMR measurements have been performed at low temperatures in $(R_xY_{1-x})Fe_2$ compounds, where R is a heavy rare-earth (Gd, Tb, Dy, Ho, Er and Tm) in the range $0 \leq x \leq 0.05$. The spectra were measured at 4.2 K, except for Dy (77.4 K). The spectra were analysed in terms of direct and indirect (through common Fe neighbors) transferred hf interactions. The direct transferred hf field amounts to a few kOe per rare earth 1st neighbor; the indirect contributions of the first two shells are smaller by a factor of ten. We have found that a model that restricts the transferred hyperfine interactions to the indirect hf field or considers the first three neighbor shells in the same way is not satisfactory to explain the changes in the hyperfine field at Y nuclei.

I. INTRODUCTION

YFe_2 is a cubic compound with the C15-type Laves phase structure; the compound orders magnetically at 542 K with preference for an [1 1 1] easy direction.[1] Measurements have shown a saturation magnetization of 2.9 μ_B per formula unit; recent work has lead to a magnetic moment of the order of -0.4 μ_B at the Y sites.[2,3] Substitution of a rare-earth R for Y does not alter the C15 structure; it leads to higher ordering temperatures and to a change of the easy direction of magnetization preferred by the corresponding RFe_2 pure compound. In case R is a heavy rare-earth, the pseudo-binary compound orders antiferromagnetically. The [89]Y-NMR spectrum of YFe_2 has a single narrow line, centered at 45.94 ± 0.02 MHz, corresponding to a 220 kOe magnetic hyperfine field. In previous work we have presented results of NMR measurements at 4.2 K in $(R_xY_{1-x})Fe_2$ for R = Tb and Ho (0.05 < x < 0.20) and at 4.2 K and 77.4 K for R = Dy (with x ≤ 0.020).[4,5] Dilution of R in Y causes a broadening of the spectra and a shift of the main peak position towards higher frequencies. The spectra with R = Tb, Ho were analysed with a model that considered only indirect effects of the rare earth atoms on the Y hyperfine field $-H_{hf}(Y)-$ through their common Fe environment: the Fe moments are affected by the rare-earth and this modifies the Y hyperfine field. Other studies in similar series have also taken into account, besides this indirect contribution, direct effects of R substitution on the hyperfine fields at R sites[6]. Other authors limited their analysis by taking into account only substitutions in the first shell of neighbors[7].

In order to investigate the presence of direct and indirect interactions and the effects of different R impurities on the hyperfine fields at Y sites in $(R_xY_{1-x})Fe_2$ we have measured [89]Y-NMR spectra at 4.2 K and 77.4 K of compounds where R is a heavy rare earth; to reduce the

possible number of components in the spectra, this work was restricted to low concentrations ($x \leq 0.05$).

II. EXPERIMENT

The compounds were prepared from high purity elements in an arc furnace under a pure argon atmosphere, and subsequently annealed at 900 $^{\circ}$C for periods of 100 hours. X-ray measurements for crystal structure analysis confirmed the desired structure. The NMR samples consisted of fine polycrystalline powder in silicone oil. For the present work we have prepared samples with $x = 0.02$ of Gd, Tb, Dy, Ho, Er and Tm, and $x = 0.05$ of Gd and Er.

Pulsed NMR spectra were measured with a SXP Bruker spectrometer and a delay line sample holder[8]; low temperatures were reached by dipping the probes directly into the cryogenic fluids. The RF pulses had widths of the order of 0.5 μs and separation of 50 μs; transverse (T_2) relaxation times at main peak positions in all spectra were of the order of milliseconds. The spectra were obtained by plotting the integral of the echo curve versus frequency.

III. EXPERIMENTAL RESULTS

Y-NMR spectra of $(R_x Y_{1-x})Fe_2$ at 4.2 K for R = Gd, Tb, Ho, Er and Tm with $x = 0.02$ are presented in Fig. 1. The amplitude of maximum echo in each spectrum is normalized to 1. At this concentration, measurements for R = Dy are characterized by relatively weaker echo signals, due to the smaller mobility of the domain walls[5]; we have found that samples with R = Er present a qualitatively similar behavior. The spectra with $x = 0.02$ show that substitution of R for Y, even at this small concentration, can generate distinct hf fields at Y nuclei, in every case exceeding the value of $H_{hf}(Y)$ in YFe_2.

Spectra with R = Gd (at 4.2 K) and Er (at 77.4 K) for $x = 0.05$ are shown in Fig. 2. Earlier measurements for R = Tb

and Ho at these concentrations have been published[4]; for R = Dy and Er, the spectra could not be obtained at 4.2 K due to the above mentioned effect.

The main peak of the spectra shows a reduction in intensity and a shift to lower frequencies, under application of an external magnetic field. Also, magnetization measurements for R = Gd, Tb, Ho and Tm at 4.2 K show a pattern of high domain wall mobility; as a consequence, the corresponding spectra at 4.2 K are attributed to signals from nuclei in domain walls. In the C15 structure, an Y site is surrounded by 4, 12 and 12 neighbor Y atoms, at distances of 0.433012, 0.707106 and 0.829156a_0, respectively, and by 12, 16 and 12 Fe atoms, at distances of 0.414578, 0.649519 and 0.819679a_0 (a_0 = 7.363 Å). An Y atom has 6, 2 and 1 Fe nearest neighbors in common with Y atoms, respectively, in the first, second and third shells of Y neighbors.

The computer analysis of the spectra assumed line intensities corresponding to a random substitution of R for Y in the three nearest shells of atomic sites around the Y atom; peaks corresponding to configurations that have a probability $\geq$ 0.01 of occurrence are included, represented by lorentzian lines. The model considers that an R substitution in one of the first three shells adds to the hyperfine field a contribution $\Delta H_{hf}(Y)$ given by

$$\Delta H_{hf}(Y) = \Delta H^d + \Delta H^i = \Delta H^d + \Delta h_j^i \cdot p_j$$

where ΔH^d and ΔH^i are the direct and indirect transferred hf terms, p_j = 6, 2 and 1 for substitutions in the 1st, 2nd and 3rd shell, respectively; Δh^i is the indirect contribution of R per Fe common neighbor in the jth shell.

The spectra with x = 0.02 are better fitted with further conditions on the model described above:
i) the transferred hf field due to R atoms in the third shell is zero; this reduces the number of relevant peaks at this concentration to five, corresponding to the

configurations (n_1, n_2) = $(0,0)$, $(0,1)$, $(0,2)$, $(1,0)$ and $(1,1)$;

ii) it is necessary to adopt distinct Δh^i parameters for the first and second shells (Δh_1^i and Δh_2^i);

iii) the only direct contribution detected was that due to one R atom in the first shell in the configuration $(1,1)$.

The same conditions allowed a satisfactory analysis of spectra with x = 0.05 for R = Gd at 4.2 K and R = Er at 77.4 K, being necessary, in addition, to introduce an indirect contribution of the third shell ($\Delta h_3^i = \Delta h_2^i$), only required for the configuration $(1,1,1)$.

Table 1 presents the values of parameters Δh_1^i, Δh_2^i and ΔH^d in MHz; for R = Er the values of ΔH^d with x = 0.02 at 4.2 K and x = 0.05 at 77.4 K are not well defined due to the additional difficulties of measurement. To convert these values from MHz to T one has to multiply by a factor 0.479.

IV. DISCUSSION

The reduction in the number of possible peaks and a higher definition of the lines has lead us to depart from the model of indirect interactions in the following points:

i) an R atom in the third shell creates a transferred hf field only if there is simultaneous occupation of the first and second shells ($n_1 = 1$ and $n_2 = 1$);

(ii) the parameter Δh_1^i (effect on Y of an R atom in the first shell via 6 Fe common neighbors, per Fe neighbor) is smaller than Δh_2^i (effect of R in the 2nd shell, per neighbor). This result can be interpreted as a possible saturation of the effect of R on the moment of its Fe nearest neighbors; (iii) the requirement of $\Delta H^d \neq 0$ for peaks $(1,1,n)$ implies that a model dependent only on the number of common Fe neighbors is not sufficient to explain the increments $\Delta H_{hf}(Y)$. On the other hand, the result shows that a minimum number of R atoms is necessary to introduce the direct interaction (parameter ΔH^d).

Our measurements indicate that the occurrence of

detectable increments in $H_{hf}(Y)$ depends on a minimum occupation number and also on the particular shell that is being occupied.

The values found for Δh^1 represent increments $\Delta H_{hf}(Y)$ in the range of 0.02 to 0.06 T . For the parameter ΔH^d, the values found at $x = 0.02$ and $x = 0.05$ correspond to a range of 0.2 to 0.35 T. Scaling hyperfine coupling factors of Campbell for Y and Ho[9], our values of Δh^1 for Y (Δh_y^1) would predict smaller values than those estimated by Al-Assadi et al[6] at Ho atoms (Δh_{Ho}^1) in compounds with R = Gd and $x \geq$ 0.10, but that would be in a same range of values ($0.05 \leq \Delta h_y^1 \leq 0.15$ T, versus $0.2 \leq \Delta h_{Ho}^1 \leq 0.4$ T). With the same scaling, our values of ΔH_y^d (0.50 to 0.88 T) fall in the range predicted for ΔH_{Ho}^d (0 to 1 T). On the other hand, our results indicate a smaller contribution from R substitution than that estimated in ref. 7; it should be noted, however, that their measurements at 77.4 K in $Gd_xY_{1-x}Fe_2$, at compositions $x \geq 0.10$, present broader spectral lines, and were analysed within a model that took into account only effects from the first shell of neighbors.

The parameters Δh_1^1, Δh_2^1 and ΔH^d show variations that are not larger than 50% along the series, from Gd to Tm. These results indicate that, at least at the present range of low concentrations, the effects on $H_{hf}(Y)$ do not depend linearly on the spin of R atoms, or on the direction of magnetization they prefer on RFe_2.

V. CONCLUSIONS

Y-NMR measurements in $(R_xY_{1-x})Fe_2$ for low concentrations have provided a detailed view of effects of R substitution in the three first shells of neighbors on $H_{hf}(Y)$. An analysis based exclusively on direct interactions or on direct and indirect contributions that treats in a qualitatively similar way the first three neighbor shells[6] is shown to be inadequate to the type of data presented here. The results show contributions that depend on the

shell where the substitution takes place and also on the number of substitutions on the other shells. The parameters do not present a large variation along the heavy R series; the direct interaction per R atom varies from 0.23 T to 0.53 T, and the indirect term from 0.017 T to 0.038 T in the first shell and 0.033 T to 0.063 T in the second shell.

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FIGURE CAPTIONS

Fig. 1 - ^{89}Y-NMR spectra of $(R_x Y_{1-x})Fe_2$ at 4.2 K for $x = 0.02$ and R = Gd, Tb, Ho, Er and Tm; the full lines are computer fits to the model described in the text.

Fig. 2 - ^{89}Y-NMR spectra for $x = 0.05$: (a) R = Er at 77.4 K and (b) R = Gd at 4.2 K; the full lines are computer fits to the model described in the text.
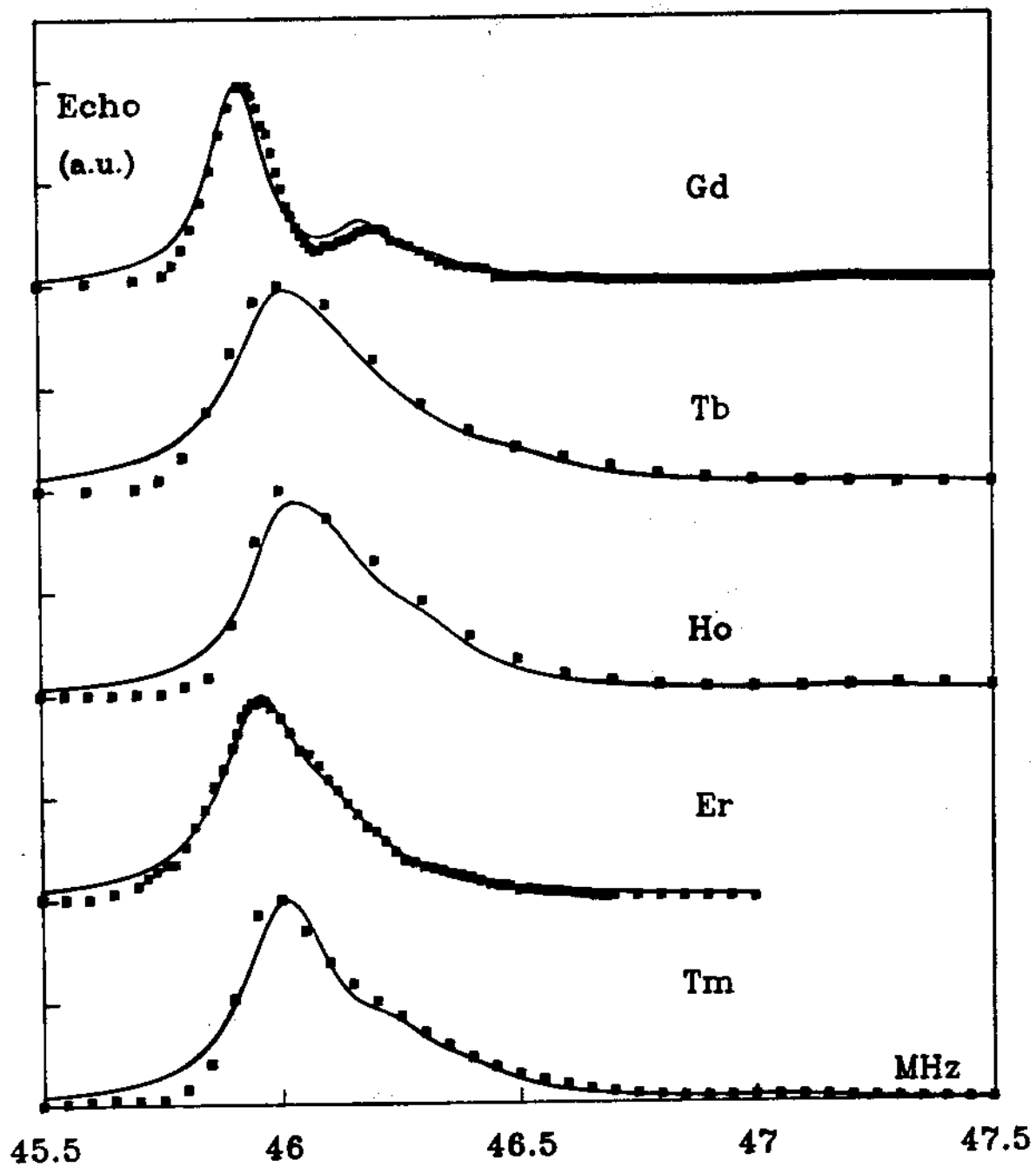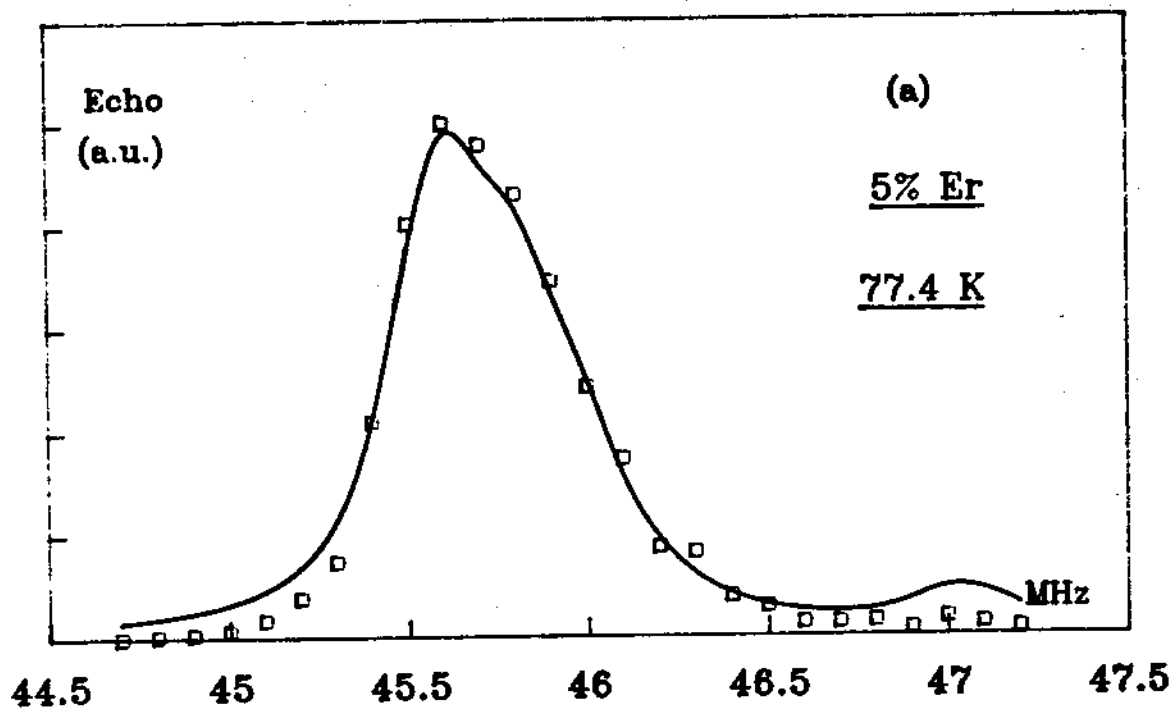
Fig. 1

Fig. 2

Tables

	Δh_1^i	Δh_2^i	ΔH^d
x = 0.02, T = 4.2 K			
Gd	0.068(1)	0.131(1)	0.610(1)
Tb	0.08(3)	0.10(2)	1.1(5)
Ho	0.050(3)	0.102(2)	0.77(5)
Er	0.036(1)	0.070(1)	1.1*
Er (77.4 K)	0.058(2)	0.08(2)	1.00(3)
Tm	0.06(2)	0.10(1)	0.48(4)
x = 0.05			
Gd (4.2 K)	0.08(2)	0.11(2)	0.6(1)
Er (77.4 K)	0.062(2)	0.10(2)	0.8*

Table 1 - Parameters Δh_1^i, Δh_2^i and ΔH^d (in MHz) in $R_1 Y_{1-x} Fe_2$; the number in parenthesis is the experimental error in the last digit. (*) parameters with additional difficulties of measurement (see text).

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