## Domain and Domain Walls NMR Spectra in RCo<sub>2</sub> Intermetallic Compounds<sup>\*</sup>

by

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#### Abstract

NMR signals in ferromagnets originate from domains and domain walls. The technique has the ability to distinguish signals originating from these two regions, through the selection of the radiofrequency (RF) power level applied to the sample. The dependence of the spin echo amplitude upon the RF field intensity can give information on magnetic properties such as domain walls mobility, distribution of local magnetic anisotropies, etc. NMR spectra obtained under different RF intensities allow the investigation of different regions of the samples. In this paper we have applied this technique to the study of the spin-echo amplitude and of the spectra of <sup>59</sup>Co in the series of the Laves phase intermetallic compounds  $RCo_2$ , where R = Gd, Dy, Tb at 4.2 K, as a function of the RF field. The results were analyzed according to a model which takes into account several features characteristic of ferromagnetic powdered samples. These include the distribution of the direction of the hyperfine field in respect to the RF field, the variation of the enhancement factor within the domain-walls, the distribution of wall areas, etc. The analysis suggests that whereas in  $TbCo_2$  the NMR signals come purely from domain walls, in  $DyCo_2$  they come mainly from domains. GdCo<sub>2</sub> behaves as an intermediate case where there exists a mixture of contributions to the NMR signals. In this case we were able to observe and measure separately the NMR spectra of nuclei from domains and domain walls.

Key-words: NMR; Domain walls; RCo<sub>2</sub>.

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## 1. Introduction

Pulse NMR has been used for the study of magnetic metals, alloys and intermetallic compounds. NMR can be applied to the study of domain walls mobility, local anisotropy, through the radiofrequency (RF) power level dependence of the spin echo amplitude signal and of the NMR spectra. In general, an NMR spectrum in a ferromagnetic system will be composed of a mixture of signals coming from domains and domain walls<sup>1</sup>. The proportion of the contributions of each of these regions to the final amplitude of the spin echo will depend on certain parameters such as the wall mobility, wall width, the local anisotropy, the exchange energy, the relative volumes of domain and domain walls, and so on. In magnetic materials the NMR signals are augmented in intensity by a quantity known as the enhancement factor. One can usually refer to domain and domain wall enhancement factors,  $\eta_D$  and  $\eta_W$ , respectively.

We study in the present paper the Laves phase intermetallic compounds  $GdCo_2$ ,  $TbCo_2$ and  $DyCo_2$  with zero-field spin echo NMR spectroscopy, and also the dependence of the spin echo amplitude with RF power level. The RCo<sub>2</sub> series (R = rare earth) is particularly suited to the purpose of the present investigation, since the magnetic anisotropy, walls width, etc., can be easily changed by replacing the rare earth R. In the analysis of the experimental results we applied a model<sup>2</sup> that includes contributions from pure domains and from domain walls to the NMR signal. In this model, the NMR signal from a ferromagnet is given by

$$\epsilon(B_1) = C_W \epsilon_W(B_1) + C_D \epsilon_D(B_1) \tag{1}$$

where  $C_W = m_o \eta_{oW} V_W$  and  $C_D = m_o \eta_D V_D$  are the relative contributions from the walls and the domains, respectively.  $V_D$  and  $V_W$  represent the fractions of the domain and domain wall volumes which effectively contribute to the signal;  $\epsilon_D(B_1)$  and  $\epsilon_W(B_1)$  are the echo intensities from domains and domain walls,  $\eta_D$  and  $\eta_{oW}$  are the enhancement factors for domains and domain wall centers.

### 2. Experimental

The intermetallic compounds were prepared by arc melting the pure elements, in an argon atmosphere; they were afterwards heat treated to ensure homogeneity. For the NMR samples the ingots were powdered and mixed with silicone oil. The measurements were made using an automated pulse NMR spectrometer and recording the spin echo amplitude; the samples were kept in a helium bath, at a temperature of 4.2 K. The excitation conditions were: pulse widths,  $\tau_a = \tau_b = \tau = 0.5 \mu s$  for GdCo<sub>2</sub> and TbCo<sub>2</sub>; for DyCo<sub>2</sub> we set  $\tau = 2.0 \mu s$ . Pulse separation  $\Delta \tau = 60 \mu s$  and repetition rate 0.1 kHz. A broadband power amplifier (ENI model 3100LA, 100 Watts) was used. The RF field ratio  $B_1/B_{1max} = 1$  corresponds to 0 dB of attenuation in the power applied to the sample.

#### 3. Results

The <sup>59</sup>Co NMR spectra consisted of one line, in the case of  $GdCo_2$  and  $DyCo_2$ ;  $TbCo_2$  presented two lines. These two lines arise from the existence of two magnetically inequiva-

lent (but crystallographically equivalent) sites which we shall label site I (lower frequency) and site II (higher frequency).

Figure 1 shows the NMR spectra of <sup>59</sup>Co in GdCo<sub>2</sub>, DyCo<sub>2</sub> and TbCo<sub>2</sub> at 4.2 K and low RF power level. The continuous lines represent Lorentzian fits from which we obtained the parameters shown in table I. Figure 2 shows the same set of spectra for maximum RF power  $(B_1/B_{1max} = 1)$ ; the respective fitted parameters are also shown in Table I. These spectra are in good agreement with reference<sup>3</sup>. We note from the table that the NMR frequencies do not change appreciably with the change in RF power in the Gd and Dy compounds. In TbCo<sub>2</sub> a shift of about 1 MHz in the first line and 2 MHz in the second line is observed. The linewidths for DyCo<sub>2</sub> and TbCo<sub>2</sub> are not much affected. However, increasing the power practically halves the linewidth of GdCo<sub>2</sub>. These features exemplify changes in NMR spectra of magnetic materials upon RF power level, mentioned at the beginning of the paper. We can try now to correlate these results with the curve of RF power in these compounds and compare with equation (1).

Figure 3 shows the curves of  $\epsilon$  vs.  $B_1/B_{1max}$  for the three compounds. The measurements were taken at the central frequencies of each spectrum, the excitation conditions as described in the previous Section, except that for DyCo<sub>2</sub> we set  $\tau = 6\mu s$ . The differences in their behavior are very clear.

In TbCo<sub>2</sub> the measurements were taken at the two magnetic sites; both curves resemble the corresponding curve for metallic iron<sup>2</sup> and are attributed to pure domain walls. Here we obtained a value of  $\eta_{oW}$  in site I which is about half of that in site II. We do not know of a similar case in the literature.

We found that a better fit of the theoretical curve to the experimental data at higher values of  $B_1/B_{1max}$  could be achieved in both lines if we superimposed another curve with  $\eta_{oW} = 1350$  in site I and  $\eta_{oW} = 1450$  in site II. This behavior could in principle be ascribed to the excitation of nuclei in the domain wall edges for large values of  $B_1/B_{1max}^{4,5}$ . In this case we would have the for same wall two different values of  $\eta_W$ : one for its center, and another for the edges. Another possibility would be to consider contributions from "50°" walls, those contained on the plane  $< 111 > \rightarrow < 11\overline{1} >$ , etc., with a smaller amplification factor. We cannot at present decide between these two possibilities, but we think two types of walls must exist in these compounds, and we tend to favor the second interpretation.

In figure 3, broken lines represent individual contributions to the final signal amplitude, whereas full lines represent equation (1).

DyCo<sub>2</sub> exhibits a situation where the maximum of the curve could just be reached for the available maximum RF power. In this figure the continuous line represents the equation that describes the NMR signals from domains, with<sup>2</sup>  $\eta_D = 8$ .

GdCo<sub>2</sub> shows an intermediate case, where we can distinguish two regions: that for  $B_1/B_{1max} \leq 0.4$  corresponding to signals from the domain walls, whereas above this value the NMR signal originates from the domains. The broken lines in the figure represent the separate contributions to the signal. Here again a better agreement between theory and experiment is achieved if we consider another curve with  $\eta_{oW} = 500$ , corresponding, e.g., to 90° domain walls (< 100 >  $\rightarrow$  < 010 >, etc.).

We can estimate the anisotropy fields in GdCo<sub>2</sub> and DyCo<sub>2</sub> using the relation<sup>6</sup>  $B_a \approx 2\pi\nu_o/\eta_D\gamma_n$ . Here  $\nu_o$  is the NMR central frequency. By replacing values from table I we

find  $B_a = 0.6$  kGauss for GdCo<sub>2</sub> and 8.2 kGauss for DyCo<sub>2</sub>, which are of the right order of magnitude<sup>4,6</sup>.

The above picture is consistent with the NMR spectral data: we should expect broader resonance lines inside the walls, since the inhomogeneity in the local field is larger in this region. The linewidths of  $DyCo_2$  (at both power levels) and  $GdCo_2$  at maximum RF power are practically the same (see table I). This fact associated with the curves of figure 3 allows us to interpret the NMR signals in  $DyCo_2$  as originating purely from the magnetic domains.

Compound	$\nu_o(\mathrm{MHz})$	$\Delta \nu (\mathrm{MHz})$	$B_1/B_{1max}$	η
$\mathrm{GdCo}_2$	61.64(7)	1.2(2)	1.0	$\eta_D = 95$
	61.75(9)	2.32(3)	0.1	$\eta_{oW}^{(180)} = 2000$
				$\eta_{oW}^{(90)} = 500$
$\mathrm{TbCo}_2$	51.39(9)	7.5(4)	0.1	$\eta_{oW}^{(I,180)} = 1800$
	61.51(3)	5.5(2)		$\eta_{oW}^{(II,180)} = 3500$
				$\eta_{oW}^{(I,50)} = 1350$
				$\eta_{oW}^{(II,50)} = 1450$
	52.5(3)	6(1)	1.0	
	63.53(7)	4.5(2)		
$\mathrm{DyCo}_2$	65.80(1)	1.13(3)	1.0	$\eta_D = 8$
	65.85(1)	1.06(4)	0.6	

Table I -  ${}^{59}$ Co NMR line positions, linewidths and enhancement factors for domain and domain walls measured in RCo<sub>2</sub> compounds at 4.2 K.

### 4. Conclusions

We have studied with pulse NMR some intermetallic compounds of the  $RCo_2$  series.

The  ${}^{59}$ Co NMR spectroscopy shows one single line for GdCo<sub>2</sub> and DyCo<sub>2</sub>, and two lines for TbCo<sub>2</sub>.

The NMR signal dependence with RF power level has been analyzed using a simple model proposed for the NMR signal intensity in magnetic materials. General agreement is obtained between this model and the experimental data on the dependence of spin echo amplitude on the RF power, supplemented by an analysis of the spectral linewidths. An effect related to the complex dependence of signals in TbCo<sub>2</sub> on the RF power level had been observed by other authors<sup>3</sup>. The origin of the frequency shift in the NMR spectra of this compound is still not clear. A similar behavior was observed in the NMR spectra of other Laves phase compounds, namely <sup>27</sup>Al in GdAl<sub>2</sub> also presenting two resonance lines<sup>7</sup>. The relatively small NMR signal in DyCo<sub>2</sub> arises from a low amplification factor in the domains, related to a large magnetic anisotropy. This compound is likely to have narrow domain walls, decreasing appreciably the parameter  $C_W$  in equation 1. This interpretation is consistent with the fact that among the three compounds studied in this paper, DyCo<sub>2</sub> is the only one where no significant changes, either in the line position or in the linewidth of the spectra, are observed with varying RF power. In GdCo<sub>2</sub> the mixture of domain and domain wall signals is more apparent. In this case, contributions from two types of domain walls (180° and 90°) seem to exist. The marked narrowing of the <sup>59</sup>Co NMR spectrum with the increase of RF power level supports the picture given here.

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### **Figure Captions**

Figure 1 - <sup>59</sup>Co NMR spectra in  $GdCo_2$ ,  $DyCo_2$  and  $TbCo_2$  for low values of the RF power, taken at 4.2 K. The continuous lines represent Lorentzian fits from which we extracted the positions and linewidths of the spectra shown in Table I.

Figure 2 - <sup>59</sup>Co NMR spectra in  $GdCo_2$ ,  $DyCo_2$  and  $TbCo_2$  at high values of the RF power, taken at 4.2 K. The continuous lines represent Lorentzian fits from which we extracted the positions and linewidths of the spectra shown in Table I.

**Figure 3** - Dependence of the spin echo amplitude on the RF power level in  $GdCo_2$ ,  $DyCo_2$  and  $TbCo_2$  at 4.2 K. The broken lines represent different contributions from domain and domain walls to the signals, and the continuous lines represent the calculated signals from equation 1.









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