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NMR AND DOMAIN WALL MOBILITY IN INTERMETALLIC COMPOUNDS*

by

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ABSTRACT: The technique of pulsed NMR can be used to study the distribution of hyperfine fields in a magnetic matrix. The dynamics of the domain walls are relevant to the generation of NMR signals. In the present study on the $(R_xY_{1-x})Fe_2$ intermetallic compounds, the reduction in the signals is associated to increased propagation fields. This indicates that a smaller domain wall mobility is at the origin of these effects. NMR spectra in this system show the importance of direct and indirect (i.e., mediated by Fe atoms) terms in the transferred hyperfine field.

Key-words: NMR; Magnetic systems.

1. THE TECHNIQUE OF PULSED NUCLEAR MAGNETIC RESONANCE (NMR)

In a nuclear magnetic resonance experiment a radio frequency field induces transitions between energy eigenstates of nuclei in a magnetic field. By recording the power absorption versus the frequency of the rf source, an NMR spectrum is obtained.

In pulsed NMR the rf is applied in pulses much shorter compared to the typical relaxation times of the system. The absorption at resonance is measured by the signal induced in a pickup coil by the precessing nuclear magnetic moments.

2. NMR IN MAGNETIC MATERIALS

In an ordered magnetic medium the NMR is affected by the interaction of the nuclei with the atomic magnetic moments; this type of resonance is sometimes called ferromagnetic nuclear resonance (FNR) (Turov and Petrov (1972), Weisman et al (1973), McCausland and Mackenzie (1979), Panissod et al (1990)). The most important consequence of this interaction is the presence of static internal fields at the nuclei, allowing the observation of zero external field NMR. The main contribution to this field is the hyperfine (hf) field; the corresponding term $\mathcal{H}_{hf}$ in the nuclear Hamiltonian is usually dominant over the interaction ($\mathcal{H}_{el}$) with the distribution of electric charges. The total Hamiltonian is given by

$$\mathcal{H} = \mathcal{H}_{hf} + \mathcal{H}_{el}$$

Several terms contribute to the hf field (see Section 5). The microscopic inhomogeneities create a distribution of hf fields, and this is responsible for the large line widths observed in FNR experiments.

The second important feature of the resonance in magnetic materials is the enhancement of the NMR signal. Since the nuclear magnetic moment and the atomic magnetic moments interact, the resonance signal is a measure of the response $\chi(\omega)$ of the coupled nuclear-atomic systems. This will be discussed in Section 2.

Finally, the nuclear-atomic interaction affects the nuclear magnetic relaxation; relaxation processes involving magnons play a dominant role.

2. INTENSITY OF THE NMR SIGNAL

2.1 Signal Enhancement

The NMR signal enhancement can be described in a simple way as consisting of two steps; first, the nuclei in a magnetic material feel an rf field that is larger than the applied $H_1(t)$ of the pulse; also, the transverse nuclear magnetization that induces a signal in the coil is magnified by the same factor. This magnification is different in domains and in domain walls.

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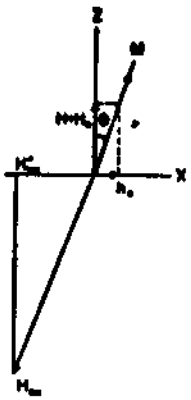

Fig. 1 Domains: H_1 tilts M, producing a large transverse component of the local (hf) field.

In the interior of the magnetic domains the enhancement arises from the transversal component of the hyperfine field that appears as the atomic magnetization is tilted an angle θ by the action of $H_1(t)$ (Fig. 1).

Since the hf field is usually very large compared to the anisotropy field, an enhancement factor η of magnitude 10 - 100 is common. η is given by

$$\eta = H_{hf}/H_a$$

where H_{hf} is the hyperfine field and H_a is the anisotropy field.

Inside a domain wall, the amplification is related to the motion of the wall due to the application of $H_1(t)$. The rf field $H_1(t)$ favors the growth of the domains that have a positive component of the magnetization along the direction of H_1; the turning of the moments in the wall generates transverse fields that are much larger than $|H_1|$ (Fig. 2).

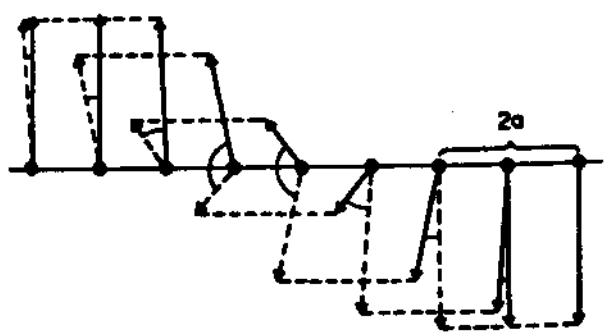

Fig. 2 Walls: H1 displaces the walls and M turns, creating large transverse hf fields.

The domain wall enhancement factor is of the order of 10^3 - 10^5 and is proportional to δx, the displacement of the wall with the rf field.

2.2. Dependence of the NMR Signal with H_1

In a pulsed NMR experiment, the nuclear magnetization is turned from the equilibrium direction (the z direction) by the application of the field H_1 in the xy plane. The resonance signal (measured, e. g. by the spin echo amplitude) is dependent on the intensity of H_1. For example, in a sequence of two pulses of equal length, the observed spin echo signal from nuclei in domain walls increases with H_1, going through a maximum (Fig. 3). The non-oscillatory behavior of the signal is due to the spatial distribution of η's, to the distribution of directions of magnetization in the different walls and to the spread in wall sizes.

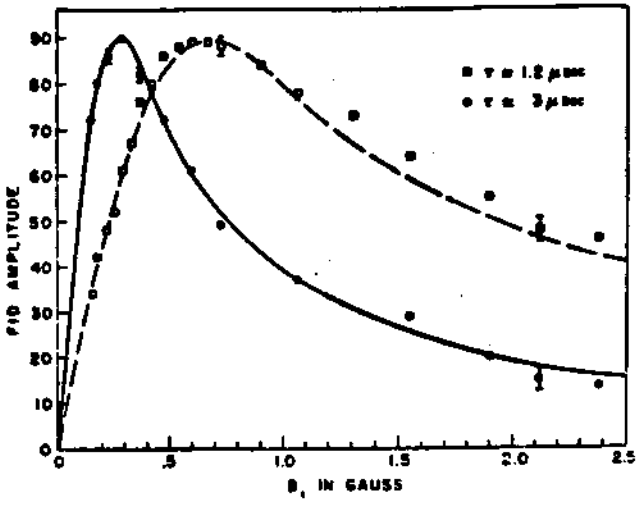

Fig. 3 NMR amplitude (FID) vs H1 in Fe; solid lines given by model (Stearns(1967)).

The experimental dependence of the intensity of the NMR signal with H_1 observed in Fe metal could be accounted for within a simple model (Stearns (1967), Stearns and Overhauser (1968)) that considers a) the average over H_1 orientations, b) a dependence of η given by $\eta(x) = C\ \mathrm{sech}(x)$; c) a model for the oscillation of domain walls (assumed circular) pinned on the edges and d)a probability distribution $p(A) \propto 1/A$ of domain wall areas. This model was also applied to nickel metal (Koster and Turrel (1969), Bohn et al (1975)).

3. MAGNETIC PROPERTIES OF THE $(R_xY_{1-x})Fe_2$ INTERMETALLIC COMPOUNDS

YFe_2 is a cubic compound with the C15-type Laves phase structure; the compound orders magnetically (e.g. Buschow (1979)) at 542 K. Y has a moment of about - 0.4 μ_B and iron of 1.7 μ_B in YFe_2 (Dumelow et al (1986)). Pseudo-binary compounds of formula $(R_xY_{1-x})Fe_2$ crystallize in the same structure. In this work we have restricted the R concentration to $x \leq 0.05$, in order to limit the number of lines in the NMR spectra.

4. INTENSITY OF NMR SIGNALS AND MAGNETIZATION

The intensity of the ^{89}Y NMR signals observed in the $(Dy_xY_{1-x})Fe_2$ compounds are much smaller than those for R = Gd, Tb, Ho, Tm (Alves et al (1986), Alves et al (1990)). The samples with Er also show a strong attenuation. The samples that show this anomaly in NMR behavior present larger values of the field where the magnetization starts to increase (propagation fields) and larger coercive fields.

One can observe that the Dy substitution for Y even in small percentages produces a reduction in the magnetic susceptibility $\chi = (\partial M/\partial H)_T$. With the data plotted in Fig. 4 for small fields, it is found that in the compound with 2% of Dy, χ is a factor of 10 smaller than the values observed in YFe_2 and in samples with R = Ho and Tb. From 2% up to 20% of Dy, the low field susceptibility suffers a reduction by another factor of 10. The coercive field H_c shows a rapid decrease with increasing temperature.

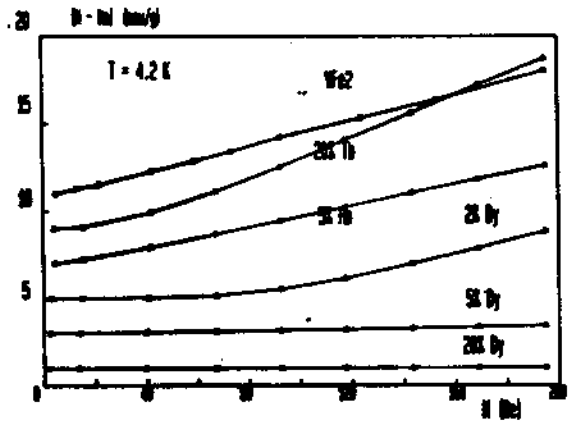

Fig. 4 Magnetization of (RY)Fe2 at low fields (4.2 K).

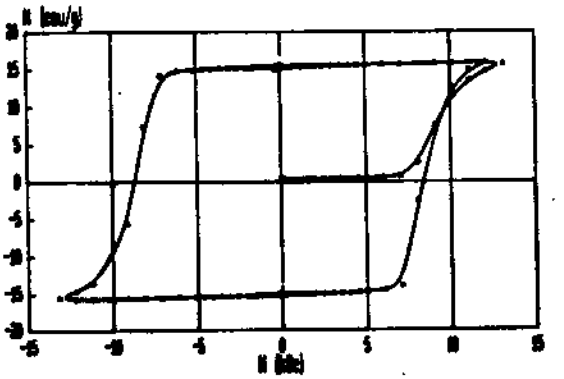

Fig. 5. Magnetization cycle for (DyY)Fe2 (x = 0.40) at 4.2 K.

The magnetization curves at 4.2 K of several samples with Dy, Tb and Ho in the concentration range $0 \leq x \leq 0.20$ are presented in Fig. 4; the figure draws attention to the initial part of the curves, corresponding to very low values of the applied magnetic field. Figure 5 shows a full cycle of magnetization for the sample with 40% Dy at 4.2 K.

The above mentioned results lead us to the conclusion that the reduction in NMR signals is attributable to a reduced mobility of the domain walls in $(R_xY_{1-x})Fe_2$, for R = Dy and Er. The difficulty in sweeping the walls

(that appears as large propagation fields) is found to be correlated to the smaller amplitude of vibration of the surface of the walls (that appears as a reduction in the echo amplitude). The common origin of these effects is attributed to the strong pinning in the presence of Dy and Er in these pseudo-binaries.

5. TRANSFERRED HYPERFINE FIELDS

The hf field in a metal consists of three main contributions: an orbital term, a core polarization term and a dipolar term. When the orbital angular moment of the atom is zero, the only non-zero contribution is the core polarization term. In addition, the magnetic hf field measured at a given nucleus that can be traced to a neighbor or to a distant magnetic moment is the transferred hf field. It may consist of a magnetic dipolar part and a conduction electron part.

The transferred hf field that acts on the nuclei through conduction electron polarization is not spatially uniform in a given matrix. If it originates from a magnetic impurity, its magnitude and sign are generally dependent on the local environment of the nucleus where the measurement is made. The NMR technique is very favorable for the observation of local variations in this field. The transferred field in a rare earth metallic system is given by (McCausland and Mackenzie (1979)):

$$\bar{H}^{n}_{tr}(A_i, \underline{r}_i) = K_n \bar{\sigma} + \sum_j f(\underline{r}_j) \langle \underline{\sigma}_j \rangle$$

where $\bar{\sigma}$ is the average spin of the alloy ($(\bar{g}-1)\langle J \rangle$), $f(\underline{r}_j)$ is a certain spatial function, and $\langle \underline{\sigma}_j \rangle$ is the spin at the impurity sites. We consider that this is the field observed at the nucleus of an atom surrounded by atoms A_i in positions $\underline{r}_i$.

In the present work, we have measured hf fields at Y nuclei in matrices of $(R_x Y_{1-x})Fe_2$; Y is very convenient for this study because the nuclei with A = 89 are 100% abundant and show no electric quadrupole interaction (I=1/2); this simplifies the analysis and leads to narrower lines. Also, Y is normally non-magnetic, and therefore the observed local magnetization

can be considered "induced". The ^{89}Y hf field in YFe_2 at low temperature is -220 kOe.

^{89}Y spin echo NMR spectra of $(R_xY_{1-x})Fe_2$ at 4.2 K for R = Gd, Tb, Ho, Er and Tm with x = 0.02 show broader lines compared to pure YFe_2; these spectra are attributed to nuclei in domain walls. At this concentration, samples with Dy exhibit weaker echo signals (Alves et al (1986)), due to the smaller mobility of the domain walls (Alves et al (1990)).

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 ≥ 0.01 of occurrence are included, represented by lorentzian lines. In the C15 structure, an Y site is surrounded by 4, 12 and 12 neighbor Y atoms, and by 12, 16 and 12 Fe atoms. 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 model considers that an R substitution in one of the first three shells contributes to the hf field with $\Delta H_{hf}(Y)$ given by

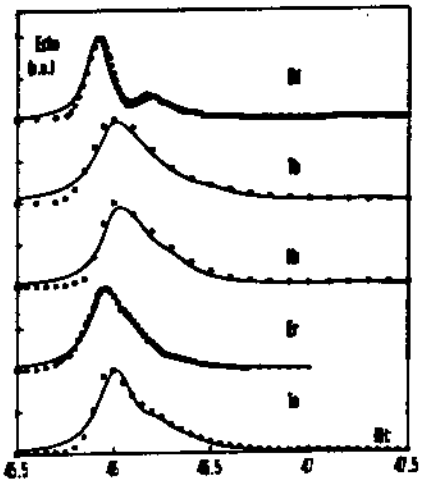

Fig. 6 - ^{89}Y NMR spectra of (RY)Fe2 (4.2 K, x = 0.02); the full lines are computer fits.

$$\Delta H_{hf}(Y) = \Delta H^d + \Delta H^i = \Delta H^d + \Delta h^i_j . p_j$$

where ΔH^d and ΔH^i are the direct and indirect (i.e. through Fe moment) transferred hf terms, p_j = 6, 2 and 1 for substitutions in the 1st, 2nd and 3rd shell, respectively; Δh^i_j is the indirect contribution of R per Fe common neighbor in the j^{th} shell (Al-Assadi et al (1984), Alves et al (1986)). The average values of the contributions for the compounds with different rare earths are: ΔH^d = 4.0 kOe, Δh^i_1 = 0.3 kOe and Δh^i_2 = 0.5 kOe.

The spectra with x = 0.02 are better fitted (Alves and Guimarães (1991)) by taking into account five configurations only: (n_1, n_2) = (0,0), (0,1), (0,2), (1,0) and (1,1); also, the direct contribution is not relevant except for R atoms in the first shell in the configuration (1,1).

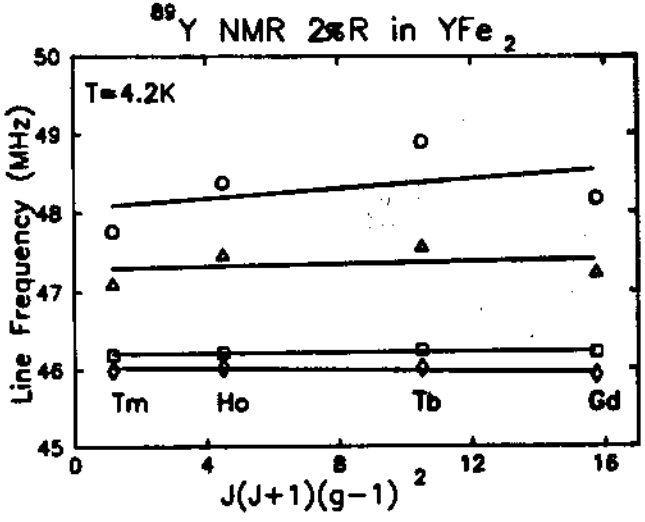

Fig. 7 - ^{89}Y NMR line positions for x = 0.02 for several R's, vs de Gennes factor.

6. CONCLUSIONS

The present study draws attention to an application of pulse NMR to the study of the magnetism of magnetically ordered materials. The intensity of NMR signals is dependent on the dynamics of domain walls; the reduced signals observed in (DyY)Fe_2 and the large propagation fields are associated to a lower mobility of such walls.

The study of the ^{89}Y hyperfine fields in these compounds reveals the presence of two kinds of transferred fields: direct (dominant), and indirect, i.e., mediated by the Fe moments.

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