Thermal Induced Resonance in (RE)Co$_2$ Magnetic Hyperfine Measurements

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Abstract: The magnetic hyperfine fields (MHF), quoted from *magnetic hyperfine interactions* (MHI) measured on the ^{140}Ce probe-isotope immersed in rare-earths/cobalt intermetallic compounds [(RE)Co$_2$] using *time-differential perturbed angular correlation* (TDPAC) technique, are presented. An excellent correlation is observed between these MHF and the evolved equilibrium temperatures at which those MHI were measured on the different compounds. A thermal induced magnetic resonance effect when $kT_m \approx g m_B B_{hf}$ is used to explain the results.

Keywords: Magnetic Hyperfine Fields; Intermetallic Compounds; Resonance

1. Introduction

The C15 Laves phases (RE)M$_2$ (RE= rare earth, M= *3d* transition metals Fe, Co, Ni) intermetallic compounds have been well studied, from both experimental and theoretical points of view, in terms of their hyperfine interaction properties (nuclear electric quadrupole and nuclear magnetic dipole). It has been shown that their magnetic properties are determined by the coexistence and interaction of the magnetic moments of the highly localized electrons of the RE-atoms and of the itinerant *3d* electrons of the transition metals [1-3]. In the case of the set of (RE)Co$_2$ compounds, the role that the electronic configuration of the isotope-probes used for *time-differential perturbed angular correlation* (TDPAC) measurements on the *magnetic hyperfine interactions* (MHI) was reviewed [1]. The purpose of this communication is to present, besides quoted magnetic hyperfine fields (MHF) obtained from TDPAC experiments on (RE)Co$_2$ compounds (RE= Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm) doped with the ^{140}Ce probe-isotope, evidence for thermal induced magnetic resonance. The details of the experimental arrangement and measurements are given below in order to simplify the understanding of the context in which the results were obtained.

2. Experimental Details

The TDPAC measurements were carried out with the 328 keV-486 keV γ-γ cascade of the $\tau_{1/2}$= 40 hs isotope ^{140}Ce populated in the β-decay of ^{140}La [^{139}La(n,γ)^{140}La $\xrightarrow{\ b\ }$ ^{140}Ce]. Samples of (RE)Co$_2$ were doped by arc-melting of the metallic reagents with ≤ 0.5 at.% of radioactive La-metal, in an argon atmosphere. The measuring systems were standard 4-detector set-ups equipped with BaF$_2$ scintillators. A detailed description of the theory and the TDPAC technique can be found in [4]. There was no specific control to ensure that all samples had the same size. Hence different sizes were used. With a characterization purpose, all the compounds were measured at room temperature (RT). The spectra (with the exception of TmCo$_2$'s which showed a small electric quadrupole interaction) displayed no interaction as inferred from the practically constant anisotropy values in large time-delay intervals. This is also evidence that ^{140}Ce was sitting on the cubic RE site of (RE)Co$_2$. The samples were cooled in closed-cycle He refrigerators where good thermal contact was established between the sample and the support cooling head. The temperatures were measured by thermo-couples attached to the samples containers. The only exception was TmCo$_2$, which was measured in a liquid-He cryostat as far as in this case the magnetic T_C is known to be lower than the minimum temperature, of about 10 K, which can be reached by the above cited He-refrigerators. The cryogenic systems were allowed to evolve freely to some final equilibrium temperature, T_m; but in one of the systems (see Table I) the lowest temperatures reached were controlled in order to avoid temperature fluctuations. These lowest reached temperatures tended to be different for the different compounds. Initially we attributed such differences to some kind of a deficiency in the cryogenic systems, which prevented the attainment of their design minimum temperatures. We now believe that T_m is strongly influenced by the sample characteristics.

3. Results and Discussion

Fig. 1 shows some of the measured spectra at the lowest temperatures, for (RE)Co$_2$, using TDPAC. The results of the fits are given in terms of the magnetic frequency obtained by:

$$\boldsymbol{u}_M = \frac{g_N \boldsymbol{m}_N B_{hf}}{h} \tag{1}$$

g_N: Landé g-factor for the nucleus; $\boldsymbol{m}_N$: nuclear Bohr magneton number; h: Planck constant. The MHF derived from these data have been submitted for a Conference presentation [5]. Table I displays the results of the internal MHF $B_{hf}(T_m)$ quoted from the measurements performed at the final equilibrium temperatures reached for each of the compound. Fig. 2 displays the variation of the internal MHF as a function of the "de Gennes factor" $(g-1)J$, where both g and J are the Landé g-factor and the total angular momentum quantum number for the RE atom respectively. Table I has been constructed

in terms of increasing atomic number of the RE atom. The MHF $B_{hf}(T_m)$ increases from PrCo$_2$ and reaches a maximum for GdCo$_2$ and then gradually drops there after.

Perhaps what also appears obvious in Table I and Fig. 2 is the apparent correlation between the quoted MHF, $B_{hf}(T_m)$, and T_m for different compounds, which can easily be assumed as a coincidence. Fig. 3 shows a plot of $B_{hf}(T_m)$ versus T_m, which shows clearly a strong correlation between the data points. The best-fit straight lines to the two sets of data are also shown in Fig. 3 and have the form

$$B_{hf}(T_m) = B_n + A_n T_m \tag{2}$$

where $n = 1, 2$ (Set 1, Set 2). The best-fit parameters for B_n and A_n are given in Table II. The correlation between $B_{hf}(T_m)$ and T_m for both sets of data is rather good. Good fits to the experimental data are obtained. We believe that these results are unlikely to be fortuitous and offer a possible explanation in terms of a thermal induced resonant effect. We also note that $A_1 \approx 2A_2$.

It has been mentioned before [6] that when the orbital magnetic moment is not completely quenched, spin-orbit coupling can split the ground state degeneracy into a multiplet of states not far apart in energy compared to thermal energy (kT). We suspect that multiplet splitting by the local magnetic field introduces sublevels with energy difference comparable to kT. The presence of RE atoms in (RE)Co$_2$ compounds also ensures the existence of strong spin-orbit coupling. The energy shifts between adjacent low-lying multiplet states can be expressed as

$$\Delta E = n g \boldsymbol{m}_B B_{eff}(T_m) \tag{3}$$

where g is the orbital g-factor, $\boldsymbol{m}_B = 9.27400899 \times 10^{-24}$ J T^{-1} is the Bohr magneton and $n = 1, 2$ (corresponding to excitations to first and second excited states). $B_{eff}(T_m)$ is the effective local field experienced by the affected electrons assumed to be

$$B_{eff}(T_m) = B_{hf}(T_m) - B_n \tag{4}$$

where B_n is a constant. It is plausible to assume that in ferromagnetic (RE)Co$_2$ compounds where there are both localized moments at RE sites and itinerant electrons from the Co, the field elsewhere might not differ very much from the field at the RE site. At most measuring temperatures the internal field will not vary very much with slight temperature changes. Hence $n g \boldsymbol{m}_B B_{eff}(T_m)$ will almost be constant except for ErCo$_2$, which has a Curie point of 30 K [1].

As the sample is being cooled slowly, we can expect a significant proportion of thermal radiation with energy coming close to that of ΔE in equation (3). When the thermal energy matches the magnetic energy splitting, resonant absorption of thermal radiation by some electrons in low lying states will occur and the system can reach equilibrium at that temperature if the rate of cooling is not that large. Under these conditions we can expect

$$n g \boldsymbol{m}_B B_{eff}(T_m) \approx k T_m \tag{5}$$

where $k = 1.3806503 \times 10^{-23}$ J K^{-1} is the Boltzmann constant. Combining equations (4) and (5) we get

$$B_{hf}(T_m) = B_n + \frac{k T_m}{n g \boldsymbol{m}_B} \tag{6}$$

Clearly a plot of $B_{hf}(T_m)$ versus T_m should be a straight line with slope,

$$Slope = \frac{k}{n g \boldsymbol{m}_B} = \frac{1.4887}{ng} \text{ T K}^{-1} \tag{7}$$

to four decimal places. This is clearly in reasonable agreement for $g \approx 1$ with the slopes of $A_1 = 1.46 \pm 0.06$ T K^{-1} and $A_2 = 0.711 \pm 0.007$ T K^{-1} obtained earlier in Fig. 3. This result appears to show that excitations of electrons between orbital L-multiplet states (for which $g_L \approx 1$). Equation (7) clearly, shows why we must expect the slopes of the plots in Fig. 3 to be as observed ($A_1 \approx 2A_2$).

The TDPAC measurements provide accurate values of the MHF. In the present results we attribute slightly more error in attaining the equilibrium temperature T_m than in $B_{hf}(T_m)$. For Set 1 (Sm, Gd, Tb, Dy, Ho, and Tm based compounds), the equilibrium temperatures observed are associated with excitations of electrons from the ground state to the first excited state ($n = 1$). Other equilibrium temperatures, corresponding to higher energy transitions can be expected at higher temperature. For Set 2 (Pr, Nd and Er based compounds), the equilibrium temperatures are associated with transitions from ground state to the second excited states ($n = 2$): lower temperatures (below our present limit) corresponding to excitation to the first excited states can be expected. Hence in principal a series of equilibrium temperature traps

$$T_{mn} = \frac{ng\textbf{m}_B B_{eff}(T_{mn})}{k} \qquad (8)$$

where $n = 1, 2, 3, \ldots$ can be found.

The approach to the equilibrium temperature at which the TDPAC measurements were performed have been shown in the current work to depend on the RE atom. However, other effects such as sample size, heat retention, radioactive isotope probe atoms and rate of cooling may also play a role. The approach to the equilibrium temperature T_m will to a large extent depend on the efficiency of the cooling system. We also expect associated small oscillations in temperature variation as T_m is approached because of the onset of repeated excitation and de-excitation processes.

4. Conclusion

We have deduced the correlation between the hyperfine fields in the (RE)Co$_2$ compounds with their equilibrium temperature in TDPAC measurement. The onset of thermal induced magnetic resonance appears to provide an adequate explanation of the observed correlation between $B_{hf}(T_m)$ and T_m. If the rate of cooling and de-excitations from higher energy level matches the rate of excitations into higher energy level from the ground state, a state of thermal equilibrium can be reached. We also note that it would be quite easy to bypass the resonant condition especially with more efficient cooling systems. The two cryogenic systems used in the present study are well configured to observe the thermal induce effects associated with multiplet splitting by the local magnetic field in the intermetallic (RE)Co$_2$ compounds.

Acknowledgements

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Table I   Magnetic Hyperfine Fields $B_{hf}(T_m)$ of (RE)Co_2 compounds measured at temperature T_m. The data is present in the order of increasing atomic number or total number 4f and 5d electrons for the RE atoms.

RE	$(g-1)J$	f+d (Electrons)	T_m (K)	$B_{hf}(T_m)$ (T)
Pr	-0.8	3	11.5*	8.12
Nd	-1.6	4	15.0	10.34
Sm	-1.786	6	10.0*	15.53
Gd	3.5	8	15.0*	22.63
Tb	3	9	13.6	21.03
Dy	2.5	10	12.0*	18.97
Ho	2	11	10.0*	16.29
Er	1.5	12	20.3	14.38
Tm	1	13	4.2	6.99

*The lowest temperatures reached were controlled in order to avoid temperature fluctuations (Hyperfine Interactions Group of IPEN/CNEN, SP/Brazil).

Table II   Parameters corresponding to the best fit straight lines in Fig. 3 corresponding to Set 1 (RE = Sm, Gd, Tb, Dy, Ho, Tm) and Set 2 (RE = Pr, Nd, Er) plots for (RE)Co_2 compounds.

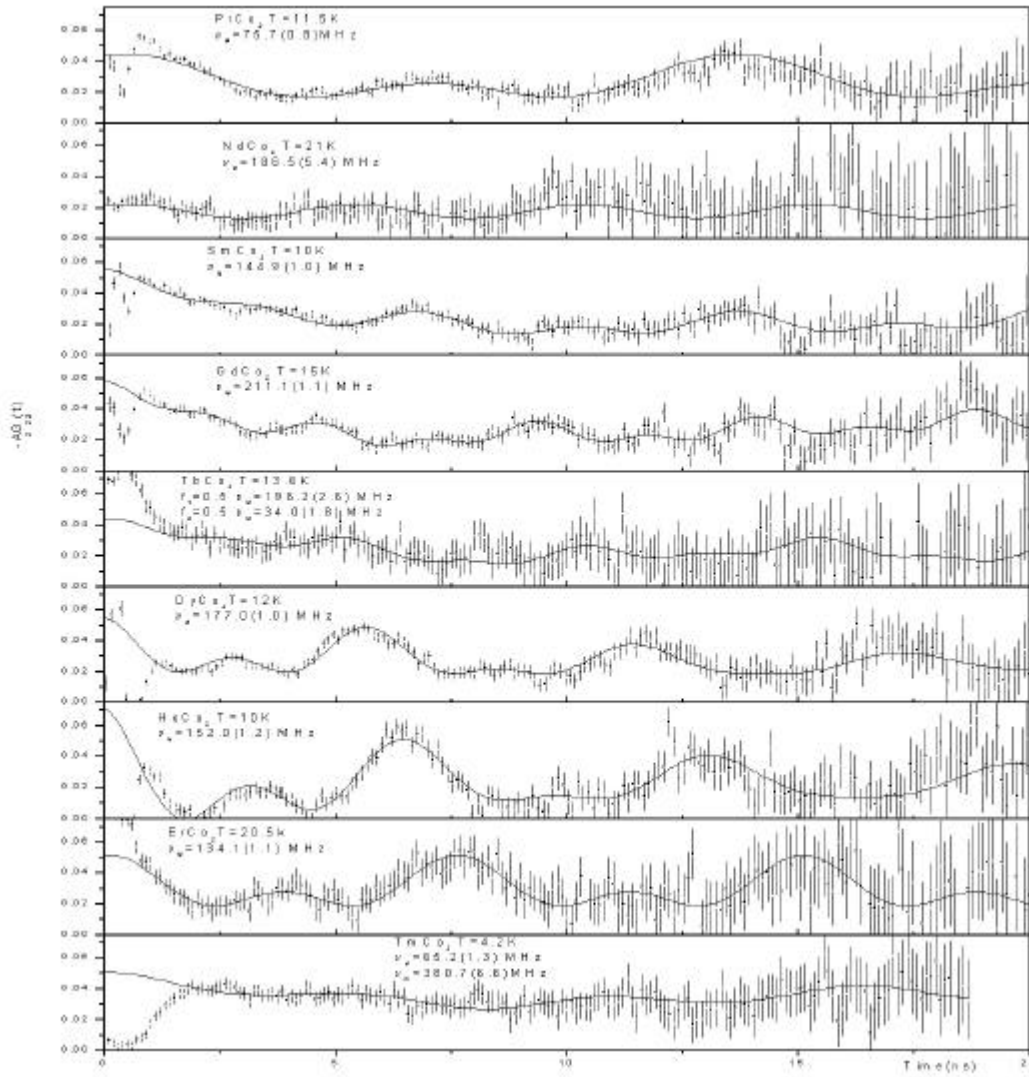| Set	B_n (T)	A_n (T K^{-1})	c^2
1	1.2 ± 0.6	1.46 ± 0.06	0.995
2	-0.07 ± 0.6	0.711 ± 0.007	0.999

Figure 1 TDPAC Measured Spectra on ^{140}Ce:(RE)Co$_2$ Compounds at Equilibrium Lowest Temperatures (see Table I for the sources of data)
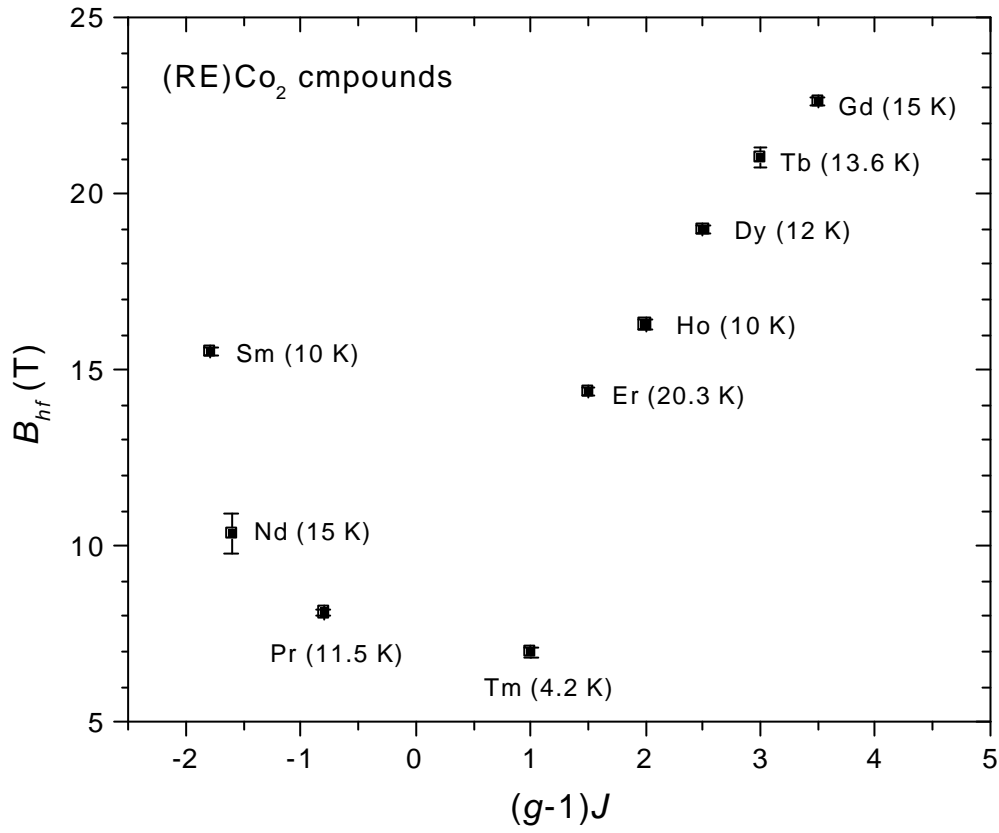
Figure 2   Magnetic Hyperfine Fields $B_{hf}(T_m)$ of (RE)Co$_2$ Compounds Plotted as Function of the "de Gennes factor" $(g-1)J$.
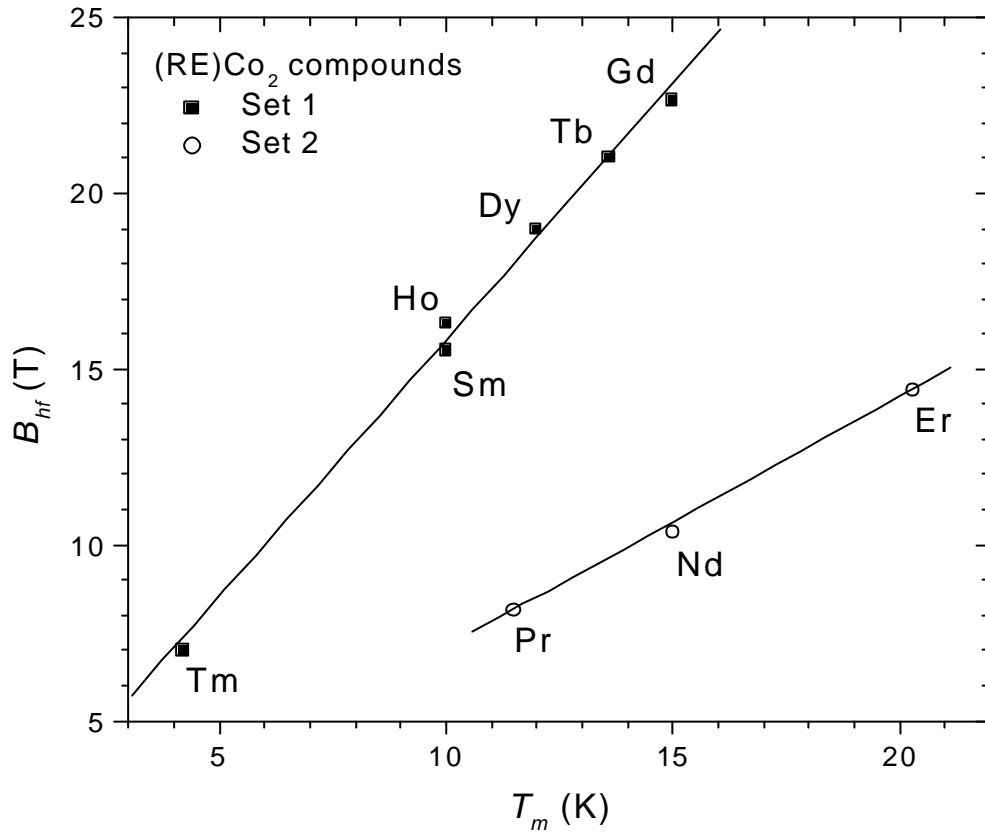
Figure 3   Magnetic Hyperfine Fields $B_{hf}(T_m)$ of (RE)Co$_2$ Compounds Plotted as Function of the Equilibrium Temperature of Measurement T_m.