2005 International Nuclear Atlantic Conference - INAC 2005
Santos, SP, Brazil, August 28 to September 2, 2005
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: 85-99141-01-5

STUDY OF MAGNETISM IN GDTIN (T = NI, PD, CU) COMPOUNDS BY PERTURBED GAMMA-GAMMA ANGULAR CORRELATION SPECTROSCOPY

D. M. T. Leite, A. L. Lapolli, A. W. Carbonari, R. N. Saxena, J. Mestnik-Filho

Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes, 2242
05508-000, São Paulo, SP, Brazil
dmtleite@ipen.br

ABSTRACT

In the present work, a nuclear technique, The Time Diferential Perturbed Gamma-gamma Angular Correlation (TDPAC) was used to measure the magnetic hyperfine field at Gd sites in the GdTIn (T = Ni, Pd, Cu) compounds using ^{140}La–^{140}Ce probe nuclei. The radioactive probe nuclei were produced by irradiation natural La with neutrons in the IEA-R1 research reactor of IPEN. Small amounts of radioactive La were arc-melted along with the constituent elements of the compounds. The crystal structure of the samples was investigated by x-ray diffraction. The PAC results showed that, below the respective magnetic ordering temperature, a pure magnetic interaction was observed at the ^{140}Ce sites for all samples. The saturation values of the magnetic hyperfine field for GdNiIn, GdPdIn and GdCuIn along with those for $GdNi_2$ and $GdCo_2$ are compared with values of the magnetic hyperfine field measured with ^{111}Cd probe nuclei for Gd compounds.

1. INTRODUCTION

The magnetic hyperfine field (mhf) at probe nuclei sites has three contributions: the orbital motion of electrons in open shells, the dipole field produced at nuclei by the spin moments of unpaired electrons, and the non-zero spin density inside the nuclear volume originated from s-character electrons. For non-magnetic closed d-shell probe nuclei, like ^{111}Cd, in a magnetic compound the only contribution to the mhf comes from the s-character electrons polarized by magnetic neighbor ions. However, for open 4f-shell probe nuclei like rare-earth elements the orbital contribution to the mhf from the electrons of the ions, which contains the probe nuclei, can be large enough to interfere in the measurement of the mhf from the magnetic ions of the compound. This problem is particularly critical when rare-earth probe nuclei are used to measure the mhf in compounds where one of the constituents is a rare-earth element. In the present work we have compared the mhf values measured with ^{140}Ce in Gd-based compounds to those measured with ^{111}Cd and have shown that in these particular compounds ^{140}Ce nuclei can probe the mhf from Gd ions without interference from their own electrons.

The samples measured in this work belong to the RTIn (R = rare-earth, T = transition metal) family of compounds, which crystallize in the Fe_2P prototype structure (hexagonal structure with space group $P\bar{6}2m$) and show interesting magnetic properties and a variety of magnetic structures [1]. This structure is formed by magnetic R–T layers alternated with non-magnetic T–X layers. The magnetic R atoms occupy the positions: x, 0, ½ ; 0, x, ½ ; $\bar{x}$, $\bar{x}$, ½ form a triangular structure, which is a deformed Kagomé lattice. One of the characteristics of such a triangular arrangement of magnetic atoms is the frustration of the magnetic interactions when

an antiferromagnetic order is present. The compounds of this family have been very little studied so far. In the present work, we have investigated the temperature dependence of the magnetic hyperfine field (B_{hf}) on Gd sites using ^{140}Ce probe sites of the GdNiIn, GdPdIn, and GdCuIn compounds.

2. EXPERIMENTAL

The polycrystalline GdTIn samples were prepared by repeated arc melting the 99,99% pure constituent elements in an arc furnace, under argon atmosphere purified with a hot titanium getterer. For each compound, radioactive ^{140}La nuclei, obtained by neutron irradiation of lanthanum metal in the IEA-R1 research reactor of IPEN, substituting about 0.1% of Gd atoms was melted along with the constituent elements. Samples were annealed under an atmosphere of ultra pure Ar for 48 h at 800 ºC. The structure of samples was checked by X-ray diffraction measurement and the experimental results were fit by the Rietveld method [2] using DBWS9807 program [3].

The Perturbed Gamma-gamma Angular Correlation (TDPAC) measurements were carried out with a conventional fast-slow coincidence set-up with four conical BaF_2 detectors. The well-known gamma cascade of 329 keV-487 keV, populated from the decay of ^{140}La, with an intermediate level with spin $I = 4^+$ at 2083 keV ($T_{1/2} = 3.45$ ns) in ^{140}Ce, was used to measure the magnetic hyperfine field (B_{hf}) at Gd sites of the samples. The samples were measured in the temperature range of 10-300 K by using a closed-cycle helium cryogenic device. The time resolution of the system was about 0.6 ns for the gamma cascade used.

The TDPAC method is based on the observation of hyperfine interaction of nuclear moments with extra-nuclear magnetic field or electric field gradient. A detailed description of the method can be found elsewhere [4,5]. The interaction between the magnetic dipole moment of the nucleus $\boldsymbol{\mu}$ and the magnetic hyperfine field $\mathbf{B}$ is given by the Hamiltonian $\mathcal{H} = \boldsymbol{\mu}\cdot\mathbf{B}$. The eigenvalues E_m are given by $E_m = -g\mu_N Bm$, where g is the nuclear g-factor and μ_N is the nuclear magneton, producing the energetically equidistant Zeeman splitting. The energy difference between two adjacent sub-levels is

$$\Delta E = E_{m+1} - E_m = \hbar\omega = g\mu_N B$$

The TDPAC method uses a radioactive nuclear probe which decays via a γ–γ cascade through an intermediate energy level with spin I which, in the presence of an external magnetic field, precesses with a Larmor frequency ω_L given by $\omega_L = g\mu_N B/\hbar$ .Spin rotation spectra $R(t)$ were generated from background subtracted coincidence counts $C(\theta,t)$,

$$R(t) = 2\left[\frac{C(180^\circ,t) - C(90^\circ,t)}{C(180^\circ,t) + 2C(90^\circ,t)}\right]$$

where $C(\theta,t)$ are the geometric mean of the coincidences taken from the spectra recorded at angle θ. The coincidence spectrum is called perturbed time-differential γ–γ angular correlation function which in polycrystalline samples is expressed (neglecting the A_{44}-terms) by the following function:

$$W(\theta,t) = 1 + A_{22}G_{22}(t)P_2(\cos\theta)$$

where θ is the angle between the detectors, A_{22} is the unperturbed angular correlation coefficient of the γ–γ cascade, $P_2(\cos\theta)$ is the Legendre polynomial and $G_{22}(t)$ is the perturbation factor. The measured perturbation function $R(t)$ was fitted by using the following model,

$$R(t) = A_{22}G_{22}(t) = A_{22}\sum_i f_i G_{22}^i(t),$$

where f_i are the fractional site population and $G_{22}^i(t)$ are the corresponding perturbation factors. In the case of magnetic dipole interaction for an unpolarized sample below the magnetic ordering temperature this factor can be written as

$$G_{22}(t) = 0.2 + 0.4\cos\omega_L t + 0.4\cos 2\omega_L t$$

The perturbation factor $G_{22}(t)$ of the correlation function contains detailed information about the hyperfine interaction. Measurement of $G_{22}(t)$ allows the determination of the Larmor frequency $\omega_L = \mu_N g B_{hf}/\hbar$, from the known g-factor of the 2083 keV state of ^{140}Ce to magnetic dipole interaction.

3. RESULTS AND DISCUSSION

The X-ray diffraction measurements indicated a single phase and Fe_2P-type structure with the $P\bar{6}2m$ space group for the samples. Some results are shown in Fig.1 and the lattice constants of all samples are included in table 1.

Table 1. Crystallographic properties of GdTIn compounds

Compound	Structure Type	Lattice constants		Reference 1	
		a (Å)	c (Å)	a (Å)	c (Å)
GdNiIn	Fe_2P	7.464	3.825	7.467 (1)	3.845 (1)
GdPdIn	Fe_2P	7.516	3.848	7.649 (1)	3.886 (1)
GdCuIn	Fe_2P	7.462	3.986	7.465	4.004

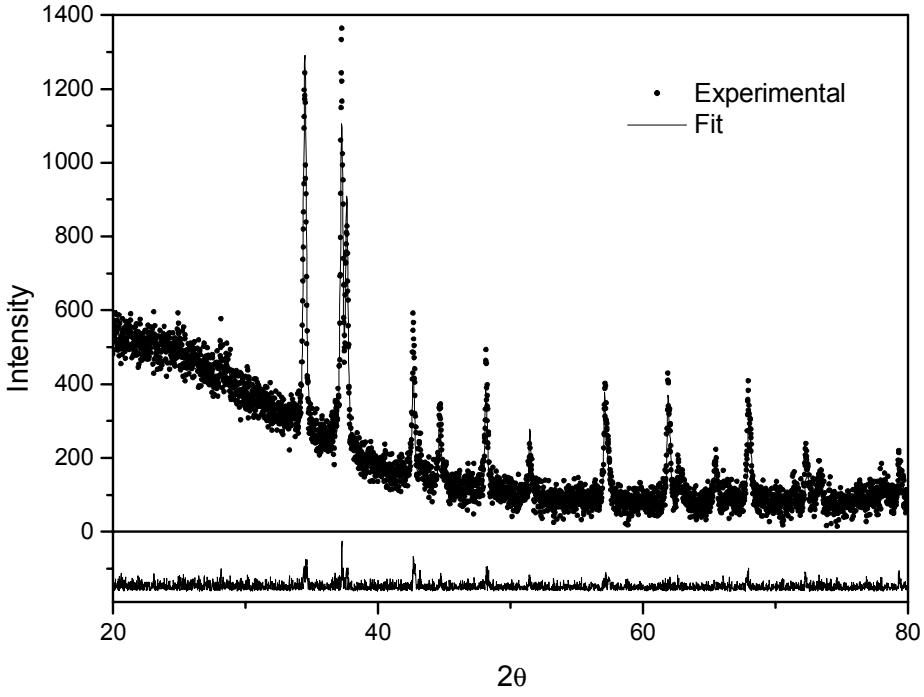

Figure 1. Results of X-ray diffraction for GdNiIn.

Some of the TDPAC spectra measured with ^{140}Ce probe nuclei are shown in Fig.2. The quadrupole moment of the 2083 keV 4^+ state of ^{140}Ce is known to be very small [6], consequently one expects to observe an almost pure magnetic dipole interaction in the antiferromagnetic phase of the sample. Below the magnetic transition temperature, a unique magnetic interaction is observed at ^{140}Ce in Gd sites of all compounds. The temperature dependence of B_{hf} for ^{140}Ce was observed to be the standard for a magnetic compound.

In reference 7, the saturation values of B_{hf} at ^{111}Cd on Gd sites in $GdNi_2$, $GdAl_2$ and Gd are compared to the respective Curie temperature and the result showed a linear dependence where B_{hf}/T_C = 0.116 T/K. In reference 8, the saturation value of 11.5 T for B_{hf} at ^{111}Cd in GdNiIn, gave a ratio of B_{hf}/T_C ~0.12 T/K and follows the same linear behavior reported in reference 7. The saturation values of B_{hf} at ^{140}Ce on Gd sites in GdNiIn, GdPdIn and GdCuIn, obtained in the present work, and in $GdNi_2$[9] and $GdCo_2$[10] are compared to the respective transition temperature of each host in Fig.3. One can observe that to a good approximation B_{hf} is a linear function of the transition temperature. According to the RKKY theory of indirect coupling the ratio between the conduction electron spin polarization (CEP) and the order temperature is expected to be proportional to $[J_{sf}(g-1)(J+1)]^{-1}$, where J_{sf} is the s-f coupling constant, g the Landé factor and J the total angular momentum. The linear relation between B_{hf} at ^{140}Ce and the magnetic transition temperature in Fig.3 thus may imply that the main contribution to the B_{hf} comes from the CEP at the probe site and the coupling constant J_{sf} has the same value in GdNiIn, $GdNi_2$ and $GdCo_2$ compounds. Therefore, the ^{140}Ce probes in this case behave as closed shell nuclei like the ^{111}Cd.

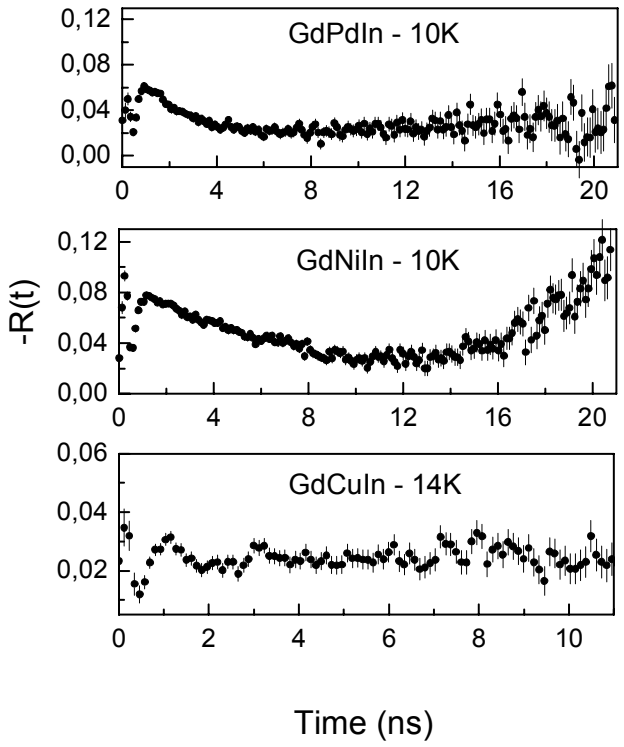

Figure 2. TDPAC spectra of the hyperfine interactions at In sites of GdPdIn, GdNiIn and GdCuIn compounds, at 15, 20 and 10 K, respectively.

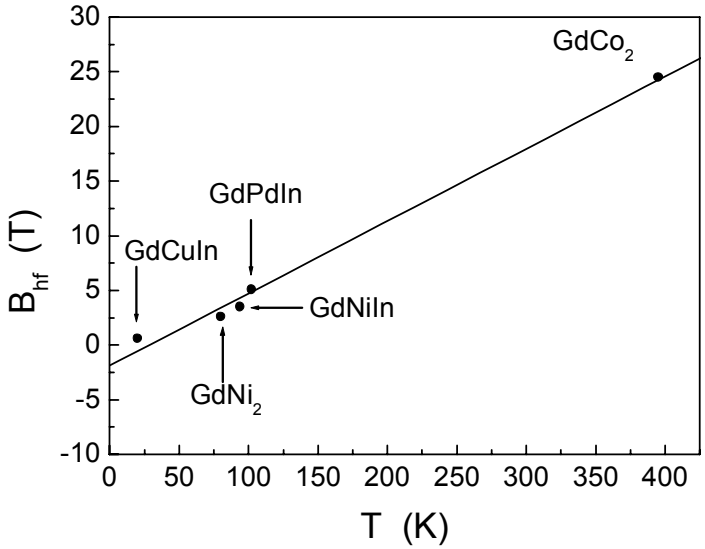

Figure 3. The saturation values of the magnetic hyperfine field B_{hf} at ^{140}Ce in some Gd compounds as a function of the respective magnetic transition temperatures.

3. CONCLUSIONS

In the present work we have used PAC technique to measure the magnetic hyperfine field at Gd sites in GdNiIn, GdPdIn and GdCuIn intermetallic compounds below their respective magnetic ordering temperature using ^{140}Ce as probe nuclei. The results have shown that ^{140}Ce nuclei, although having one unpaired f electron, can be used to probe magnetic hyperfine field at Gd sites in some Gd compounds as well as a non-magnetic probe.

ACKNOWLEDGMENTS

Partial financial support for this research was provided by the Fundação de Amparo para Pesquisa do Estado de São Paulo (FAPESP). D. M. T. Leite acknowledges the student fellowship granted by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

REFERENCES

[1] P. Villars , A. Prince , H. Okamoto, *Handbook of ternary alloy phase diagrams*, ASM International, Materials Park, OH (1991).
[2] H. M. Rietveld, "A profile refinement method for nuclear and magnetic structures", *J. Appl. Cryst.*, v. 2, pp. 65-71 (1969).
[3] D. B. Wiles, R. A. Young, "A new computer program for Rietveld analyses of X-ray powder diffraction patterns", *J. Appl. Cry*st., **14**, pp.149-151 (1998).
[4] A. W. Carbonari, R. N. Saxena, W. Pendl Jr., J. Mestnik Filho, R. Attili, M. Olzon-Dionysio, S. D. de Souza. *J. Mag. Mag. Material*, **163,** p.p. 313 (1996).
[5] R. Dogra, A. C. Junqueira, R.N. Saxena, A.W. Carbonari, J. Mestnik-Filho, M. Morales, *Phys. Rev. B*, **63**, p.p. 224104 (2001).
[6] K. Królas, B. Wodniecka, H. Niewodniczanski Institute of Nuclear Physics, Kraków, Poland, Report No. 1644/OS-1993 (unpublished).
[7] S. Müller, P. de La Presa, M. Forker, *Hyperfine Interactions*, **133,** p.p. 59 (2001).
[8] A. L. Lapolli, A. W. Carbonari, R. N. Saxena, J. Mestnik-Filho to be published in Hyperfine Interactions.
[9] A. W. Carbonari, private communication.
[10] J. Mestnik-Filho, H. Saitovitch, P. R. J. Silva, A.W. Carbonari, to be published in Hyperfine Interactions.