2011 International Nuclear Atlantic Conference - INAC 2011
Belo Horizonte,MG, Brazil, October 24-28, 2011
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
***ISBN:* 978-85-99141-04-5**

Magnetic hyperfine field in TbZn compound measured by PAC spectroscopy using ^{111}Cd and ^{140}Ce as probe nuclei

Bosch-Santos B., Cabrera-Pasca G. A., Carbonari A.W., Costa, M. S., Saxena R. N.

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

ABSTRACT

The ferromagnetic compound TbZn has been investigated by perturbed γ-γ angular correlation (PAC) spectroscopy using $^{111}In \rightarrow ^{111}Cd$ ($t_{1/2}$ = 85 ns) and $^{140}La \rightarrow ^{140}Ce$ ($t_{1/2}$ = 3,4 ns) as probe nuclei. Measurements were carried out in the temperature range of 10 – 295K for the ^{111}Cd probe and below Curie temperature (T_C) for the ^{140}Ce probe. The study of magnetic properties of rare earth (R) and zinc compounds of the type RZn is interesting because Zn ion is a closed shell atom and the rare earth elements present a localized magnetism associated with 4*f* electrons, which do not participate in chemical bonds. Therefore, the magnetic properties of these compounds are only originated from the 4*f* electrons of rare earth. Furthermore, the compounds exhibit the highly symmetric cubic structure of the CsCl prototype and have a Curie temperature of TbZn is T_C ~ 204 K [1].

1. INTRODUCTION

As mentioned earlier, the study of magnetic properties of rare earth and zinc (RZn) compounds is interesting because the rare earth elements present a localized magnetism associated with 4*f* electrons, which do not participate in chemical bonds, and are responsible for the magnetic properties in these compounds. The TbZn compound ferromagnetic exhibits a cubic crystal structure CsCl type belong to space group Pm-3m and with a T_C ~ 204 K (Curie temperature) [1]. A tetragonal distortion below Curie temperature was observed for the TbZn compound. The saturation moment at 4.2 K lies along the [110] and its value is 8.85 μ_B [2]. It is reported that when this compound is formed in the sample TbZn also appears a small percentage of the Tb metal. Terbium exhibits a hexagonal crystal structure belong to space group P63/mmc with a T_C = 219K and a Neel temperature, T_N = 230K. The TbZn compound represents a prefer orient along [110] direction below 63K whereas above this temperature the Tb magnetic moments prefer orient along [001] direction [3].
The PAC spectroscopy is based on the emission of two gamma radiations in a cascade arising from the decay of the excited states of a probe nucleus and provides information about the hyperfine interaction between nuclear external fields and nuclear moments of probe nuclei at a certain atomic site in the crystalline structure of the compound, and consequently allows extracting information of the involved hyperfine parameters, as well as the characterization of structural and magnetic transitions of the crystal. The PAC measurements were carried out using a conventional fast-slow coincidence set-up using four conical BaF_2 detectors (figure 1). The gamma cascade of 172 – 245 keV populated from the decay of ^{111}In with an intermediate level with spin I = $5/2^+$ at 245 keV ($T_{1/2}$ = 84.5 ns) in ^{111}Cd, was used to investigate the hyperfine interactions at In sites. The gamma cascade of 329 – 487 keV populated from the decay of ^{140}La with an intermediate level with spin I = 4^+ at 2083 keV ($T_{1/2}$ = 3.4 ns) in ^{140}Ce.

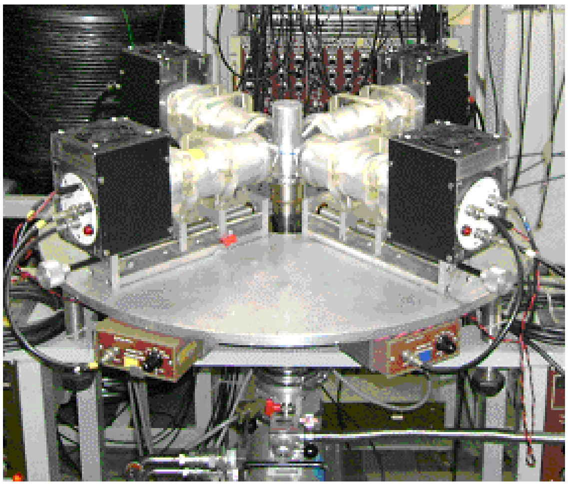


Figure 1. PAC Spectrometer with four BaF_2 detectors.

^{111}In (^{111}Cd) probe presents a high quadrupole moment (Q = 0.83 b [4]) so the electric quadrupole interaction can be seen. ^{140}La (^{140}Ce) probe is very interesting for the PAC technique because it show low quadrupole moment (Q = 0.35b [4]) and the quadrupole frequency cannot be observed, what allows only measurements of the MHF.

2. EXPERIMENTAL

Samples of TbZn compound (Tb = 99.9% e Zn = 99.999% purity) were prepared by arc-melting in stoichiometric proportions, and were characterized by X – ray diffraction (figure 2). The results showed a major fraction of the sample corresponding to the expected CsCl structure and a minor fraction corresponding to the hexagonal structure of the metal Tb. Carrier-free ^{111}In (^{111}Cd) probe nuclei were diffused into the samples at 800 ºC in vacuum. ^{140}La (^{140}Ce) probe nuclei were added to the sample by arc-melting it along with a small piece of natural La irradiated with neutrons in the IEA-R1 research reactor of IPEN followed by a thermal treatment. PAC measurements were carried out in the temperature range of 10 K to 295 K.

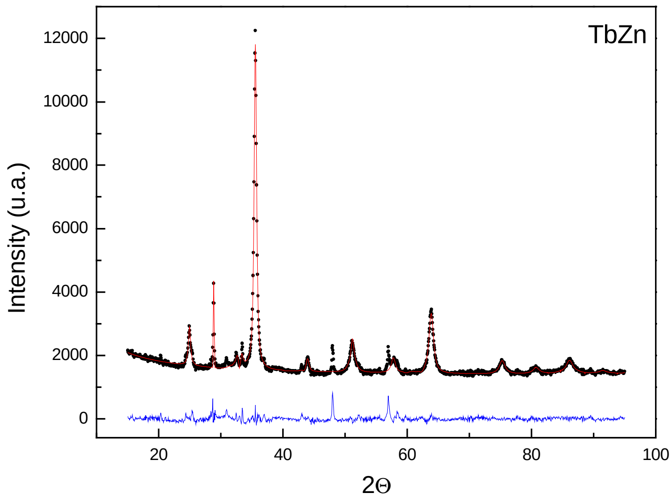


Figure 2. X-ray diffraction TbZn compound. Presents two phases, main phase is cubic structure of TbZn and the second phase corresponding to hexagonal structure of Tb.

Measurements were taken in the temperature ranges of 10 – 295 K and 10 – 200 K for ^{111}In-^{111}Cd and ^{140}La-^{140}Ce probes, respectively. ^{140}La (^{140}Ce) probe were obtained by the irradiation of natural La with neutrons in the IEA-R1 reactor at IPEN research reactor with a neutron flux around 3 x 10^{13} n/cm^2-s for a period of 10 hours. ^{140}La nuclei were added to the samples by arc-melting them again along with a small piece of irradiated natural La followed by a thermal treatment at 750 ^{0}C for 12 hours sealed in vacuum. ^{111}In was diffused into the samples which was sealed in vacuum and annealed at 700 ºC for 8 hours.
PAC measurements yields twelve coincidence spectra W(θ,t) that were analyzed by means of TDPAC software [5] and it output the $A_{22}G_{22}(t)$ function, this function contains detailed information about the hyperfine interaction and allows the determination of Larmor frequency (equation 1) and observe the electric quadrupole frequency.

$$\omega_L = -g\frac{\mu_N}{\hbar}\vec{B}_z \qquad (1)$$

Thus, g is the g-factor of intermediate state, μ_N is the nuclear magneton, $\hbar$ is the Planck constant and B_Z is the magnetic field. From the know g-factor, g = (0.306 ± 0.001) of the 245 keV state of ^{111}Cd and the g-factor of ^{140}Ce probe is g = (1.014 ± 0.038) [6], it is thus possible to determine the magnetic hyperfine field B_{hf}.

3. RESULTS AND DISCUSSIONS

After the thermal diffusion of the compound with probe nuclei the sample is taken to the PAC spectrometer, and a first measurement is performed at room temperature for checked if the compound is in the correct structure and annealing was efficient for the thermal diffusion of the probe nuclei. In the case ^{111}Cd probe nuclei, the main findings were both electric and magnetic interactions and for the ^{140}Ce probe nuclei only magnetic interaction were found

because for these probe nuclei the quadrupole moment (Q) of the intermediate state involved in the gamma cascade is very small.
The PAC spectra for ^{111}Cd probe in TbZn are shown in figure 3, the measurement were carried out in the temperature range 10 – 295 K. Below T_C these results show one site with magnetic and electric quadrupole interactions. The results of PAC spectra show two sites, both have a magnetic frequency and an electric quadrupole frequency. The main site have a minor magnetic frequency and the major fraction 65% approximately, corresponds to probe nuclei replaced in the atomic position of cubic structure of TbZn (site 1) and the other site have a major magnetic frequency and minor fraction aproximately 35%, corresponds to probe nuclei replaced atomic position of hexagonal structure of Tb (site 2).

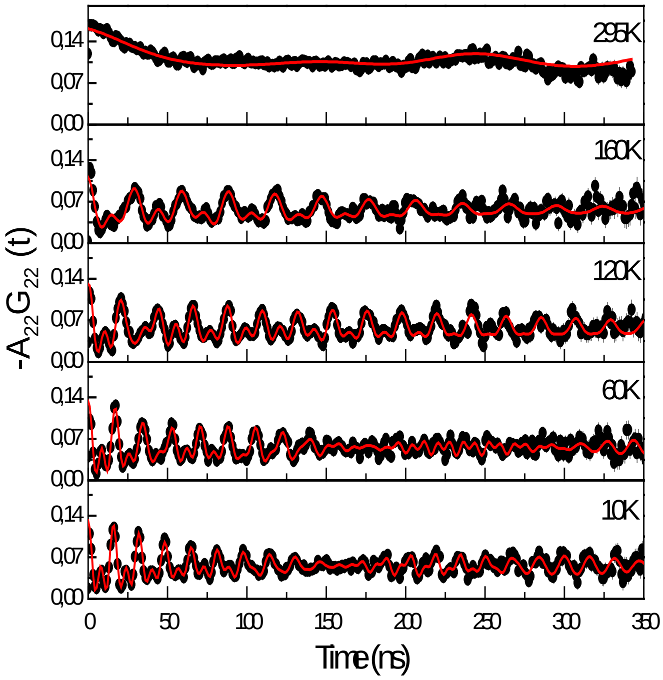


Figure 3 – PAC spectra for ^{111}In(^{111}Cd) probe in TbZn at different temperatures. The solid lines are the least square fit to the theoretical function.

The sites could be associated through value of Curie temperature, in the case TbZn (T_C = 204K), as Figure 4, the magnetic frequency goes to zero close this temperature. For the case of Tb, the magnetic frequency goes to zero near the 219K (Néel temperature). The hyperfine parameters at 10 K were for site 1 (TbZn), $\nu_{M1} = (61,67 \pm 0,05)$ *MHz*, $\nu_{Q1} = (6,87 \pm 0,16)$ *MHz*, $\delta_1 = (0,85 \pm 0,14)$ *%*, and for site 2 (Tb), $\nu_{M2} = (65,02 \pm 0,05)$ *MHz*, $\nu_{Q2} = (15,14 \pm 0,28)$ *MHz*, $\delta_2 = (0,60 \pm 0,09)$ *%*.

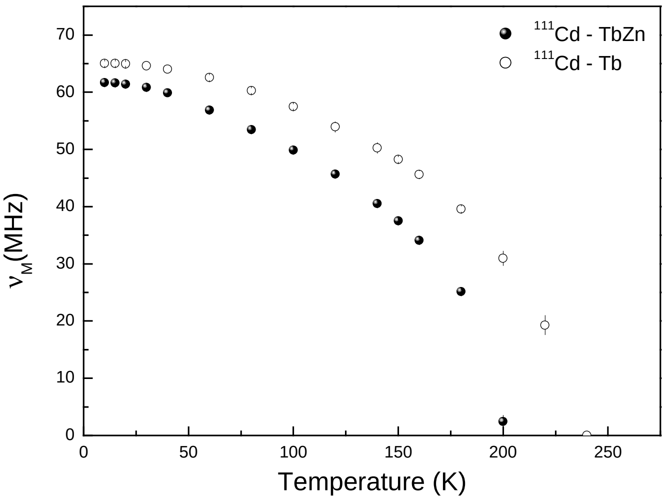


Figure 4 – Temperature dependence of the magnetic frequency for TbZn dopped ^{111}Cd probe.

Considering ^{140}Ce probe the PAC spectra are show in figure 5, was looked only one site corresponding to the probe nuclei replacing a position in the cubic structure TbZn. Thus the hyperfine parameters at 10K were ν_M = *(96,97 ± 8,63) MHz*, δ = *(0,27 ± 0,05).* For the case 40K, where is the maximum value for frequency, the parameters hyperfine were ν_M = *(241,01 ± 4,31) MHz*, δ = *(0,13 ± 0,01)*.

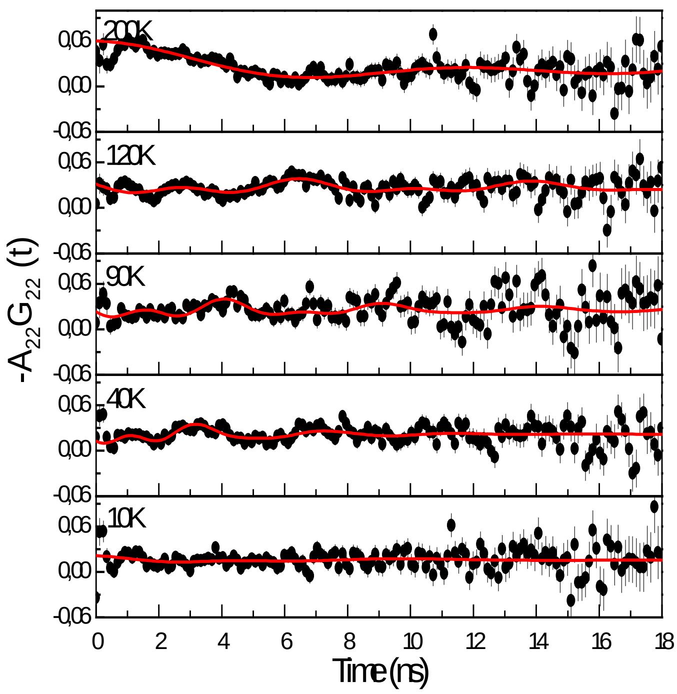
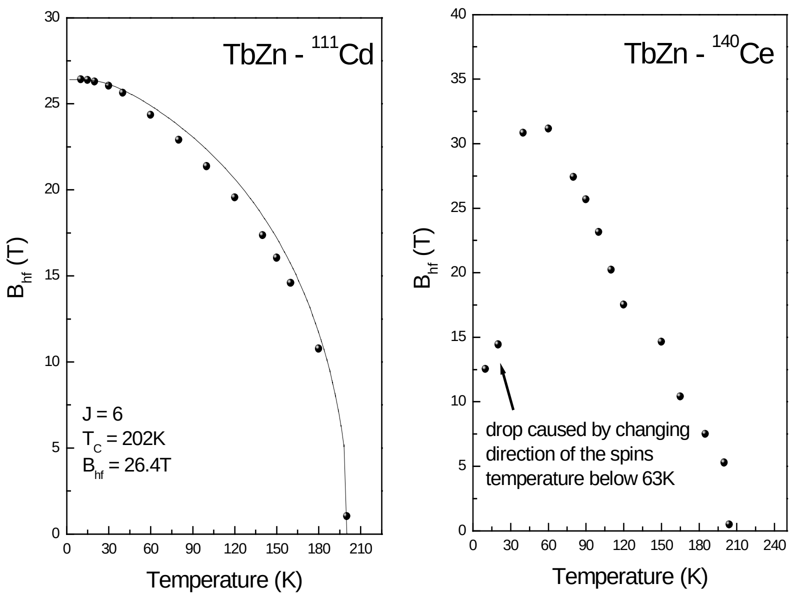


Figure 5 – PAC spectra for ^{140}La(^{140}Ce) probe in TbZn at some temperatures. The solid lines are the least square fit to the theoretical function.

The figure 5 show anomalous behavior of the Brillouin function and also evidences that the magnetic frequency reaches a maximum at 60K then falls again. This decrease in the frequency can occur because the moments of the TbZn compound are positioned themselves along the direction [001], but below 63K the moments prefer to orient along the [110], then this change in the direction of the moments can cause this decrease in the value of the magnetic frequency [7].

A similar behavior was seen in PAC measurement of the magnetic hyperfine field depends on the time to ^{140}Ce in Tb in the article by Thiel et al. [7]. The tests showed that at temperatures below 30K the hyperfine magnetic interaction decreases allowing conclude that there is an admixture of a non-magnetic state which increases at lower temperatures and a proposed descrition for this behavior was that of the non-magnetic state can be incorporated into the model of Jaccarino. Thus, this behavior of the frequency can also be caused by a mixture of a non magnetic temperature increases below 60K.

Thus, with Larmor frequency (equation 1) was calculated magnetic hyperfine field (mhf) for the compound dopped with ^{111}Cd and ^{140}Ce nuclei probes. For the two cases plotted the temperature dependence of the magnetic hyperfine field (Figure 6). For the case ^{111}Cd nuceli probe was fitted the Brillouin curve for J_{Tb} = 7/2, in this fit was observed the Curie temperature of 202K being in the range of variation found other references.



(A) (B)

Figure 6 – A) Temperature dependence of the ordered magnetic moment for (^{111}Cd) probe in TbZn. The solid line corresponds to the Brillouin function for J = 6. B) The temperature dependence of the magnetic hyperfine field of ^{140}Ce in TbZn. It is observed a probable structural transition.

PAC measurements performed with ^{111}In-^{111}Cd probes showed an electric quadrupole interaction above T_C and a combined electric quadrupole plus magnetic interaction below T_C that has been assigned to ^{111}Cd probes replace Zn atoms because has a better affinity than with rare earth atoms. In the case of ^{140}Ce probes nuclei, PAC measurements below T_C

showed only a magnetic interaction which was assigned to ^{140}La probe replace Tb atom positions.

3. CONCLUSIONS

The results show the existence two fractions for measurements with ^{111}Cd probe nuclei. The major fraction with lower Larmor frequency, which showed to be temperature dependent below T_C, has been assigned to probe nuclei replacing Zn sites in the cubic structure of TbZn. The temperature dependence of the magnetic hyperfine field which emerged from this fraction was fitted by a Brillouin function for J = 6. The higher frequency associated to the minor fraction corresponds to probe nuclei substituting the Tb sites in the hexagonal structure of Tb (T_C= 219K e T_N= 230K). Results for the magnetic hyperfine field with ^{140}Ce probe nuclei show also two similar fractions. However, the temperature dependence of the magnetic hyperfine field correspondent to the major fraction showed a sharp deviation from an expected standard Brillouin function and a mixture of a magnetic with a non magnetic state below 30K.

ACKNOWLEDGMENTS

Partial financial support for this research was provided by the Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

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