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Study of the magnetic properties of GdZn compound using PAC spectroscopy with ^{140}Ce and ^{111}Cd as probe nuclei

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Abstract In this work, we have investigated magnetic properties of the ferromagnetic GdZn compound by perturbed γ–γ angular correlation (PAC) spectroscopy using ^{140}La→^{140}Ce and ^{111}In→^{111}Cd as probe nuclei. Measurements were carried out in the temperature range of 10–300 K for the ^{111}Cd probe and below Curie temperature (T_C) for the ^{140}Ce probe. GdZn compound exhibits the cubic structure of CsCl prototype and orders ferromagnetically at $T_C = 268$ K. PAC measurements taken 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}In probes substituing Zn 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 substituting Gd atom positions. The results for the measured magnetic hyperfine field are discussed in terms of the spin polarization of the magnetic ions.

Keywords Rare-earth magnetism · Gd-based compounds · Magnetic hyperfine field · Perturbed angular correlation

1 Introduction

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 4f electrons, which do not participate in chemical bonds, and are responsible for the magnetic properties in these compounds [1]. The PAC spectroscopy pro-

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vides informations about the hyperfine interactions between nuclear external fields and nuclear moments of a probe nucleus at a certain atomic site in a crystalline structure, and consequently allows extracting information of the involved hyperfine parameters, as well as the characterization of phase transitions of the crystal [2]. This technique is based on the emission of two gamma radiations in a cascade as a result from the decay of the excited states of the probe nucleus. ^{111}In(^{111}Cd) probe presents a high quadrupole moment (Q = 0.83 b [3]) 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 = 0.35 b [3]) and the quadrupole frequency can not be observed, what allows only measurements of the MHF. The GdZn compound ferromagnetic exhibits a cubic crystal structure CsCl type belong to space group Pm-3m and with a T_C= 268 K. A tetragonal distortion below Curie temperature was observed for the GdZn compound. The saturation moment at 4.2 K lies along the [101] and its value is 7.30 μ_B [1].

2 Experimental procedure

The sample of GdZn (Gd = 99.9% e Zn = 99.999% purity) was prepared by arc-melting the constituent elements in estoiquiometric proportions. Samples have been characterized by X-ray diffraction measurements, and it was observed a major fraction corresponding to the phase of the GdZn structure with Pm-3m space group. The PAC measurements were carried out using a conventional fast-slow coincidence set-up using four conical BaF_2 detectors. Measurements were taken in the temperature ranges of 10–300 K and 10–270 K for ^{111}In-^{111}Cd and ^{140}La-^{140}Ce probes, respectively. 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. ^{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 3×10^{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 800°C for 6 hours sealed in vacuum. ^{111}In was diffused into the samples which was sealed in vacuum and annealed at 800°C for 6 hours.

The perturbation factor G_{22}(t) of correlation function contains detailed information about the hyperfine interaction. Measurement of G_{22} (t) allows the determination of Larmor frequency $\omega_L = -g\frac{\mu_N}{\eta}\vec{B}_z$. From the known g-factor, g = (0.306 ± 0.001) of the 245 keV state of ^{111}Cd and the g-factor of ^{140}Ce probe g = (1.014 ± 0.038) [4], it is thus possible to determine the magnetic hyperfine field B_{hf}.

3 Results and discussion

The PAC spectra for ^{111}Cd probe in GdZn measured at indicated temperature are shown in Fig. 1a. Results for ^{111}Cd probe nuclei above the Curie temperature show a major site with pure electric quadrupole interaction with a very small quadrupole

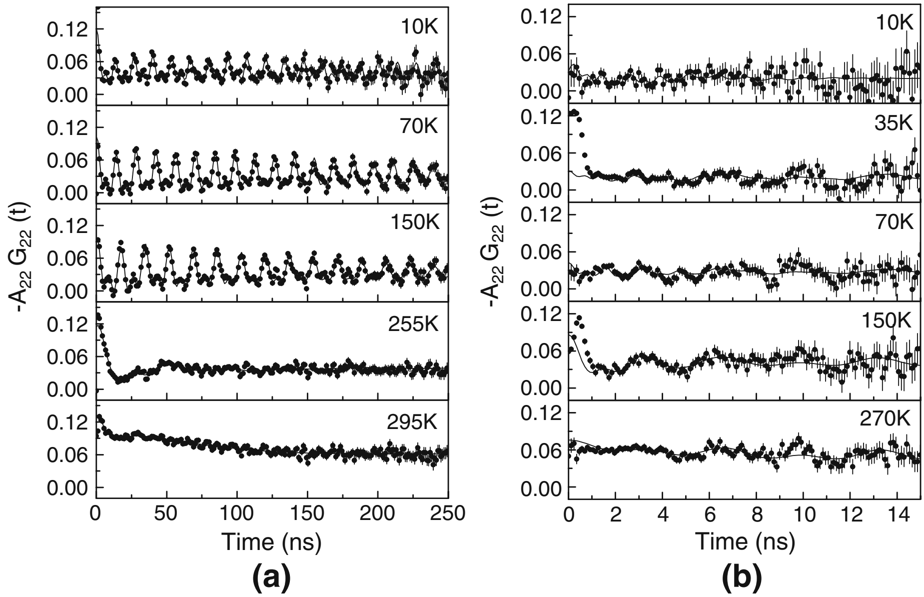

Fig. 1 **a** PAC spectra for ^{111}In(^{111}Cd) probe in GdZn at different temperatures. **b** PAC spectra for ^{140}La(^{140}Ce) probe in GdZn at indicated temperatures. The solid lines are the least square fit to the theoretical function

frequency, as expected for a cubic crystalline structure. Spectra taken below T_C using ^{111}In-^{111}Cd probes show for the major site a combined electric and magnetic interactions while in the case ^{140}Ce probe nuclei only magnetic interaction were observed. This is because for ^{140}Ce probe nuclei the small value of the quadrupole moment and the short half-life (which determine the time window for the observation of quadrupole frequencies) of the intermediate state involved in the gamma cascade impose that only quadrupole interactions with frequencies much higher than those usually found in most materials are observed.

In the case of ^{111}Cd-GdZn measurements it was observed one major frequency which was assigned to probe nuclei replacing a regular position of Zn atom in the GdZn cubic structure. The hyperfine parameters at 10 K were $\nu_M = (74.8 \pm 0.1)$ MHz, $\nu_Q = (1.6 \pm 0.9)$ MHz and $\delta = (0.5 \pm 0.2)$ %.

Figure 1b shows the PAC spectra for ^{140}La(^{140}Ce) probe in GdZn at some temperatures within the measured temperature range. Below T_C the results show two sites with dipole magnetic frequencies. One of the sites, which fraction is approximately (29.2 ± 3.5) % at 10 K, have been assigned to ^{140}Ce nuclei substituting Gd positions in the GdZn structure with dipole magnetic frequency $\nu_M = (775.3 \pm 5.2)$ MHz and frequency distribution $\delta = (3 \pm 1)$ % also at 10 K. A second dipole magnetic frequency has also been observe however, independent on the temperature. This frequency is more likely due to probe nuclei at non-substitutional sites, and, as it was observed in other PAC experiments using ^{140}Ce as probe nuclei, one possible explanation would be ^{140}Ce at clusters of La-^{140}La.

In the case of ^{111}Cd-GdZn, the temperature dependence of the magnetic hyperfine field (B_{hf}) at Zn sites can be fitted by the Brillouin function for $J = 7/2$ of the Gd ion,

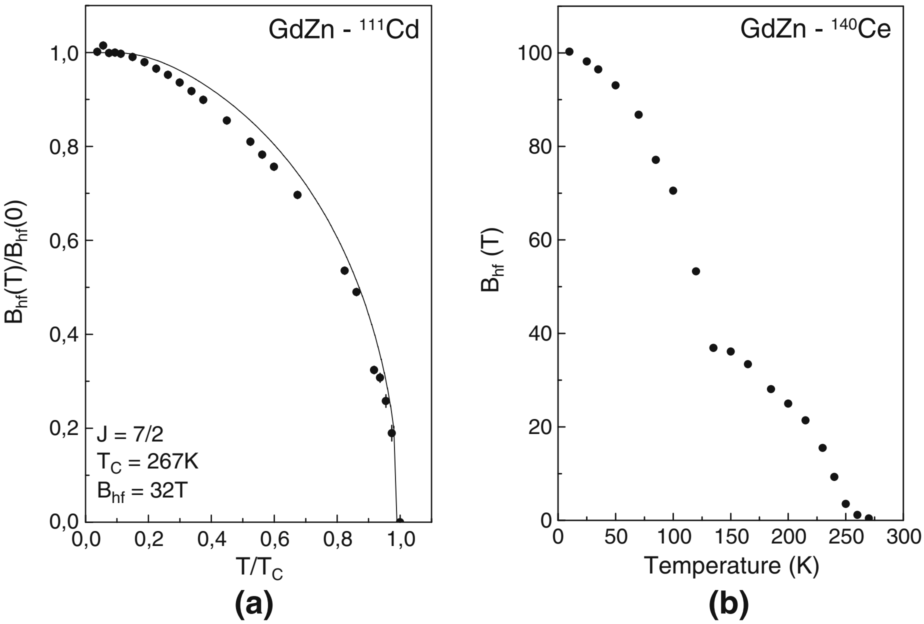

Fig. 2 **a** Reduced temperature dependence of the reduced B_{hf} for ^{111}Cd probe in GdZn. The solid line corresponds to the Brillouin function for $J = 7/2$. **b** The temperature dependence of B_{hf} for ^{140}Ce in GdZn

as shown in Fig. 2a, which means that in GdZn the magnetic exchange interaction is mediated by conduction electrons. The fitting of a Brillouin function yields a magnetic hyperfine field $B_{hf} = 32$ T at zero Kelvin and a transition temperature $T_C = 267$ K. The temperature dependence of B_{hf} measured with ^{140}Ce shows an unusual behavior (Fig. 2b), which could not be fitted by a Brillouin function. Previous works where the temperature dependence of B_{hf} was also measured with ^{140}Ce probe nuclei have also shown quite similar behavior [5–7]. The measured magnetic hyperfine field at 10 K is $B_{hf} = 100.3$ T.

Kasamatsu et al. [8] measured the MHF for GdZn and GdCd compounds with NMR measurements. Their results for GdZn are $B_{hf} = -37$ T for ^{115}In probe nuclei, and $B_{hf} = 9.89$ T for ^{139}La. In the case GdCd compound the results were $B_{hf} = -31.8$ T for ^{111}Cd probe and $B_{hf} = 10.7$ T for ^{139}La. In another work, Kasamatsu et al. [9] observed that with NMR measurements using ^{139}La as probe nuclei in GdZn $B_{hf} = 10.5$ T, and for GdCd the result was $B_{hf} = 11.3$ T. Knowing that these two compounds present the same CsCl type crystal structure, the MHF values found by these authors can be compared to those found in this work. The MHF measured with ^{115}In for GdZn and measured with ^{111}Cd for GdCd are consistent with PAC results for GdZn with ^{111}Cd probe ($B_{hf} = 32$ T). However the MHF found by Kasamatsu et al. [8, 9] with ^{139}La for both compounds is much smaller than those found by the PAC measurements with ^{140}Ce ($B_{hf} = 100.3$ T), which indicate that probably the ^{140}Ce probe is not in the valence state Ce^{4+} (no 4f electrons) but the state of Ce ions is near to 3+ (one 4f electron) and, thus, there is a contribution of the 4f electron to the measured B_{hf}. This contribution to the total magnetic hyperfine field comes from the orbital momentum of the 4*f* electron.

4 Conclusions

Results for PAC measurements of the MHF at ^{111}Cd probes in GdZn indicate the presence of only one fraction, and the temperature dependence of the B_{hf} can be fitted by the Brillouin function for $J = 7/2$. However, the ^{140}Ce-GdZn measurements showed a sharp deviation from an expected standard Brillouin function. This deviation behavior occurs due to the fact that the interaction "host" – probe nuclei is very strong, and the 4*f* electron in ^{140}Ce is spin polarized by the host field, resulting in a contribution from the probe nuclei to the total hyperfine field.

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