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ScienceDirect
Physica B 389 (2007) 168–171
PHYSICA B
www.elsevier.com/locate/physb

Investigation of the magnetic hyperfine field at Gd and In sites in GdTIn (T = Ni, Pd, Cu) compounds

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Abstract

Perturbed gamma–gamma angular correlation technique was used to measure the hyperfine interactions in the magnetic compounds GdNiIn, GdPdIn and GdCuIn using the $^{111}In \rightarrow {}^{111}Cd$ and $^{140}La \rightarrow {}^{140}Ce$ probe nuclei at the In and Gd sites, respectively. In each compound and for the measurements above the magnetic ordering temperature a unique quadrupole frequency with high asymmetry parameter η was observed for ^{111}Cd probe at In sites. Below the ordering temperature, the spectra for ^{111}Cd show a combined magnetic dipole plus electric quadrupole interaction whereas a unique magnetic interaction was observed for ^{140}Ce spectra. A linear relationship between the saturated magnetic hyperfine field (MHF) and the magnetic transition temperature was observed for both probes, indicating that the main contribution to the MHF comes from the conduction electron polarization.

PACS: 76.80.+y; 75.50.−y; 31.30.Gs

Keywords: Magnetic hyperfine field; Quadrupole interaction; Rare earth magnetism; PAC spectroscopy

1. Introduction

The RTIn (R = rare earth metal, T = transition metal) family of compounds that crystallize in the ZrNiAl-prototype structure has been investigated very little so far. The structure of such compounds is formed by magnetic R–T layers alternated with non-magnetic T–In layers. The magnetic R atoms occupy the positions: x, 0, $\frac{1}{2}$; 0, x, $\frac{1}{2}$; $\bar{x}$, $\bar{x}$, $\frac{1}{2}$ forming a triangular structure, which is a deformed Kagomé lattice. One feature of such a triangular arrangement of magnetic atoms is the frustration of the magnetic interactions when an antiferromagnetic order is present. In this family, GdNiIn and GdPdIn order ferromagnetically below 96 K [1] and 102 K [2], respectively, while GdCuIn was reported to order antiferromagnetically below 20 K [3]. Some magnetic anomalies were observed in this family, for instance in GdNiIn, Canepa et al. [4] in measurements of the Magnetocaloric properties observed a ferromagnetic transition at 96 K, and Tyvanchuk et al. [5], in AC and DC bulk magnetic measurements reported an anomalous behavior at 80 K. It was suggested that because RNiIn compounds are good electrical conductor and since interatomic distance between R atoms is large, the observed magnetic ordering would be caused by interactions via conduction electrons described by the RKKY model. The origin of the coupling mechanism between Gd moments responsible for such a difference in the magnetic behavior of these compounds is, however, not fully understood yet.

In the present work, we investigate the local magnetism by measuring the magnetic hyperfine field (MHF) on both Gd and In sites using ^{140}Ce and ^{111}Cd probe nuclei, respectively with the perturbed gamma–gamma angular correlation (PAC) technique.

2. Experimental

The polycrystalline samples were prepared by repeatedly melting pure constituent elements in an arc furnace under argon atmosphere purified with a hot titanium getterer. Carrier-free ^{111}In nuclei were introduced into the sample by thermal diffusion. For each compound, another sample was

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doi:10.1016/j.physb.2006.07.048

prepared in a similar way but with radioactive ^{140}La nuclei, obtained by neutron irradiation of lanthanum metal in the IEA-R1 research reactor at IPEN. The probe substitutes about 0.1% of Gd atoms melted along with the constituent elements. Samples were annealed under an atmosphere of ultra pure Ar for 48 h at 800 °C. The structure of the samples was checked by X-ray diffraction measurements, which indicated a single phase and ZrNiAl-type structure with the P$\bar{6}$2m space group for each compound.

The perturbed gamma–gamma angular correlation (TDPAC) measurements of the samples were carried out with a conventional fast–slow coincidence set-up with four conical BaF_2 detectors. The well-known 329–487 keV gamma–gamma cascade in ^{140}Ce populated by the beta decay of ^{140}La, with an intermediate spin level $I = 4^+$ at 2083 keV ($T_{1/2} = 3.45$ ns), was used to measure the magnetic hyperfine field (B_{hf}) at Gd sites. The gamma cascade of 172–245 keV, populated from the decay of ^{111}In with an intermediate level spin $I = 5/2^+$ at 245 keV ($T_{1/2} = 84.5$ ns) in ^{111}Cd, was used to investigate B_{hf} at In sites in GdTIn samples.

The samples were measured in the temperature range of 10–300 K using a closed-cycle helium cryogenic device. The time resolution of the system was about 0.6 ns for the gamma cascades used. A detailed description of the PAC method as well as the experimental procedure can be found elsewhere [6,7].

3. Results and discussion

Some of the TDPAC spectra measured at low temperature with ^{140}Ce and ^{111}Cd probe nuclei are shown in Fig. 1 for each alloy. In each compound the PAC results for ^{111}Cd show well-defined quadrupole frequencies above the respective magnetic transition temperature. Below the ordering temperature the measurements showed a combined interaction at the In site and a pure magnetic interaction at the Gd sites. The quadrupole moment of the 2083 keV 4^+ state of ^{140}Ce is known to be very small [8], consequently one expects to observe an almost pure magnetic dipole interaction in the antiferromagnetic phase of the sample. Below the respective magnetic ordering temperatures, unique magnetic interactions are observed at ^{140}Ce at Gd sites in all the three compounds. The temperature dependence of B_{hf} for ^{140}Ce and ^{111}Cd are shown in Fig. 2. One can observe that there is a slight increase in the B_{hf} values at low temperature where the hyperfine field should be saturated. It is not clear if this increase is real because the B_{hf} values are quite low and, consequently, less precise.

The TDPAC spectra for ^{111}Cd in GdNiIn at room temperature shows a unique quadrupole interaction with a sharp frequency $\nu_Q = 82.9(3)$ MHz, $\delta = 1\%$ and $\eta = 0.78(5)$. Below 60 K, the spectra for ^{111}Cd show combined magnetic dipole plus electric quadrupole interaction. All these spectra are characterized by a single quadrupole frequency ($\nu_Q = 84$ MHz) with $\delta = 1\%$ and $\eta = 0.78$, and a temperature dependent magnetic dipole interaction shown in Fig. 1. The angle between the EFG and MHF changes from 60° at 50 K to 30° at 20 K. A comparison of the temperature dependence of the B_{hf} for both probes shows a significant difference in the transition temperature, around 55 and 85 K for ^{111}Cd and ^{140}Ce, respectively. We have no explanation for this difference yet. The values of the MHF at low temperature for ^{111}Cd

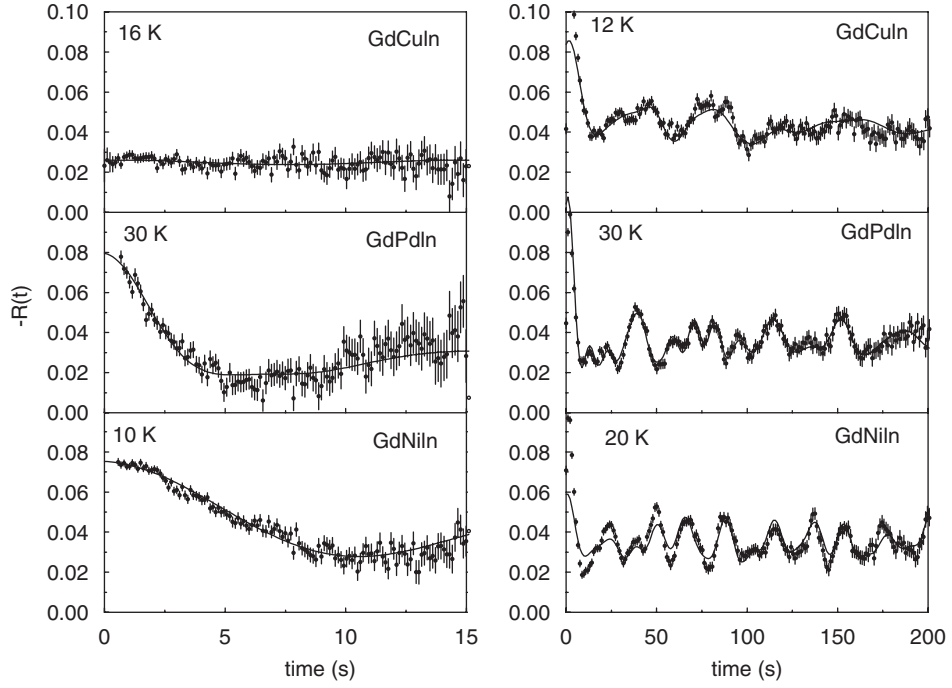

Fig. 1. TDPAC spectra at indicated temperature for ^{140}Ce (right) and ^{111}Cd (left) probes, respectively.

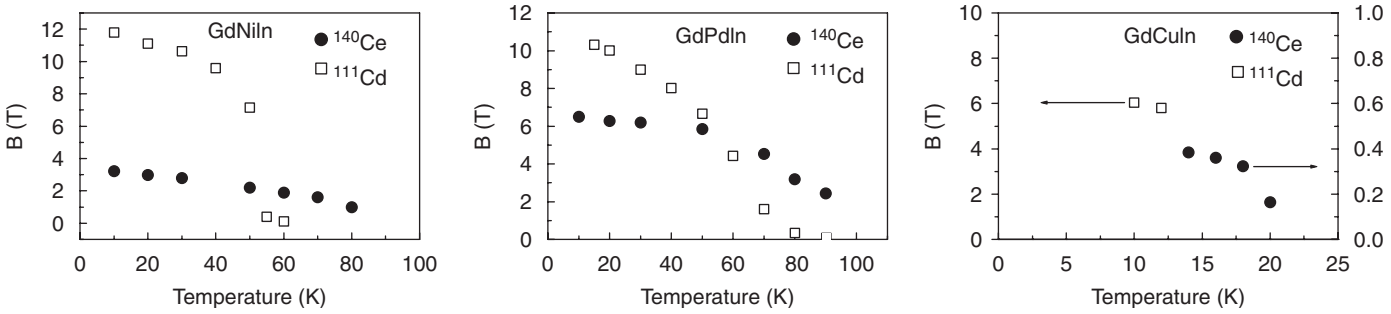

Fig. 2. Temperature dependence of the magnetic hyperfine field at In and Gd sites in the samples for ^{111}Cd and ^{140}Ce probes, respectively.

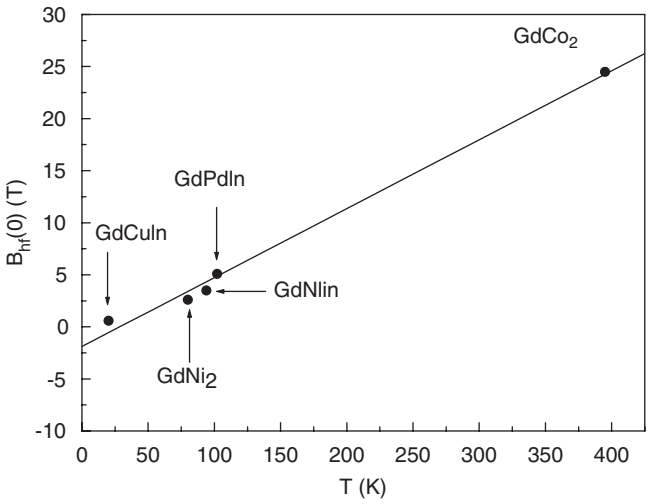

Fig. 3. The extrapolated magnetic hyperfine field B_{hf} (0) at ^{140}Ce in some Gd compounds as a function of the respective magnetic transition temperatures.

are four times larger than the values for ^{140}Ce for GdNiIn and almost two times larger for GdPdIn. This fact is mainly due to a difference in the distances of the probes to the magnetic Gd ion (~3.2 Å for ^{111}Cd–Gd and ~3.9 Å for ^{140}Ce–Gd). The saturation values of B_{hf} at ^{111}Cd on Gd sites in $GdNi_2$, $GdAl_2$ and pure Gd were compared to the respective Curie temperature [9] and the result showed a linear dependence with $B_{hf}/T_C = 0.116$ T/K. Using 11.5 and 12 T as the saturation values for B_{hf} at ^{111}Cd in GdNiIn and GdPdIn, respectively, the ratio $B_{hf}/T_C = 0.12$ T/K and follows the same linear behavior reported in Ref. [9]. The ratio for GdCuIn is two times higher. The saturation values of B_{hf} at ^{140}Ce on Gd sites in GdNiIn, GdPdIn, GdCuIn, $GdNi_2$ [10], and $GdCo_2$ [11] 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 ordering 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 seen 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 behaves as closed shell nuclei like ^{111}Cd. Preliminary ab initio calculations using the WIEN2K code have shown that the main contribution to the MHF in GdNiIn using ^{111}Cd as impurity at In sites comes from valence electrons. In Ref. [3] the magnetic hyperfine field for ^{155}Gd at Gd sites in GdNiIn, GdPdIn and GdCuIn was measured by Mössbauer spectroscopy and the results at 4.2 K are 23.3, 24 and 26.2 T, respectively. These values are five times higher than those observed for ^{140}Ce at Gd sites in the same compounds. Even if we take into account the difference in the contribution for the outer s electrons [12,13] in ^{155}Gd and ^{140}Ce, the differences would not be so high. The main contribution to the B_{hf} in ^{155}Gd comes from the core polarization, which must be much smaller in ^{140}Ce. Then, we conclude that the B_{hf} observed at ^{140}Ce in Gd sites of GdTIn alloys is mainly due to the conduction electron polarization by the Gd neighbors.

Acknowledgments

Partial financial support for this research was provided by the Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP). AWC and RNS thank CNPq for the research fellowship grant.

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