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Magnetic hyperfine fields at Gd and in sites in GdPdIn compound

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Abstract Perturbed gamma-gamma angular correlation (PAC) technique was used to measure the hyperfine interactions in the intermetallic compound GdPdIn using ^{111}In→ ^{111}Cd and ^{140}La→ ^{140}Ce probe nuclei at the In and Gd sites, respectively. The PAC results for ^{111}Cd show two well-defined electric quadrupole frequencies above T_C assigned to probes occupying Gd and In sites, with ∼50% of site occupation each. The fraction corresponding to In sites increases with temperature reaching 95% around 500 K. Below T_C the measurements for ^{111}Cd probe showed combined electric quadrupole plus magnetic dipole interaction with sharp increase in the magnetic field below around 80 K. A pure magnetic interaction with lower hyperfine field values was observed at the Gd sites occupied by ^{140}Ce below 100 K.

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

1 Introduction

Ternary intermetallic compounds of the type RMX, where R is a rare earth metal, M a transition metal and X an sp element form a series of compounds with different types of structures with interesting magnetic properties. One of such a series of compound crystallizes in the ZrNiAl-type hexagonal structure with $P\bar{6}2m$ space group. In this type of structure magnetic rare earth atoms form magnetic layers R-M separated by the non magnetic layers M-X. The rare earth atoms in each layer form triangular structure which is a deformed Kagomé lattice. One of the characterstics of this type of structure is frustration of the magnetic interaction if rare earth

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moments couple antiferromagnetically. The intermetallic compound GdPdIn orders ferromagnetically below around 102 K [1]. In the present work, local magnetism in GdPdIn has been investigated by measuring the magnetic hyperfine field (mhf) on both Gd and In sites using ^{140}Ce and ^{111}Cd probe nuclei, respectively using the perturbed gamma-gamma angular correlation (PAC) technique.

2 Experimental

The polycrystalline samples of GdPdIn were prepared by repeatedly melting pure constituent elements (Pd-99.999%, In-99.9999% and Gd-99.9%) 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 at 1,000 K for 72 h followed by slow cooling. Another sample was prepared in a similar manner but with radioactive ^{140}La nuclei, substituting about 0.1% of Gd atoms melted along with the constituent elements. ^{140}La was produced by irradiating about 1 mg of La metal with thermal neutrons in the IEA-R1 reactor at IPEN. The PAC measurements were carried out in the temperature range of 10–1,000 K. A detailed description of PAC method as well as the experimental procedure can be found elsewhere [2, 3].

3 Results and discussion

The PAC spectra for ^{111}Cd (see results at some temperatures in Fig. 1) measured above T_C show two distinct quadrupole interactions with sharp frequencies, for example: ν_{Q1} = 47.5 MHz, δ_1 = 5% and η_1 = 0.2 and ν_{Q2} = 33.0 MHz, δ_2 = 9% and η_2 = 0.6 measured at 150 K. The site populations were found to be almost equal at this temperature. First-principle calculations of electric field gradients [4] suggest that ν_{Q2} should be assigned to ^{111}Cd occupying In sites. We have no explanation for the location of the probe nuclei corresponding to the higher observed frequency ν_{Q1}. Possibilities are that this frequencies can be assigned to ^{111}Cd probes occupying In sites in another phase where the constituent elements have formed a different compound, or ^{111}Cd probes trapped in defects like vacancies and probes occupy Gd sites in GdPdIn compound. PAC measurements on binary compounds containing rare-earth and sp elements have shown that ^{111}Cd probe can occupy rare-earth as well as sp-elements [5, 6]. Below 80 K, the spectra for ^{111}Cd occupying In sites show combined magnetic dipole plus electric quadrupole interaction characterized by a single quadrupole frequency and a temperature dependent magnetic dipole interaction. The angle between the efg and mhf changes from 60° at 50 K to 30° at 20 K.

The quadrupole moment of the 2,083 keV 4^+ state of ^{140}Ce is known to be very small, consequently one expects to observe an almost pure magnetic dipole interaction in the ferromagnetic phase of the sample, measured with this probe. The temperature dependence of B_{hf} for ^{140}Ce and ^{111}Cd are shown in Fig. 2 along with the Brillouin curve for J = 7/2 that was fitted to the experimental data in order to determine the saturation values of B_{hf} at 0 K and the Curie temperatures. A comparison of the temperature dependence of the B_{hf} for ^{111}Cd and ^{140}Ce probes shows significantly different transition temperatures,with the values of about 76 K

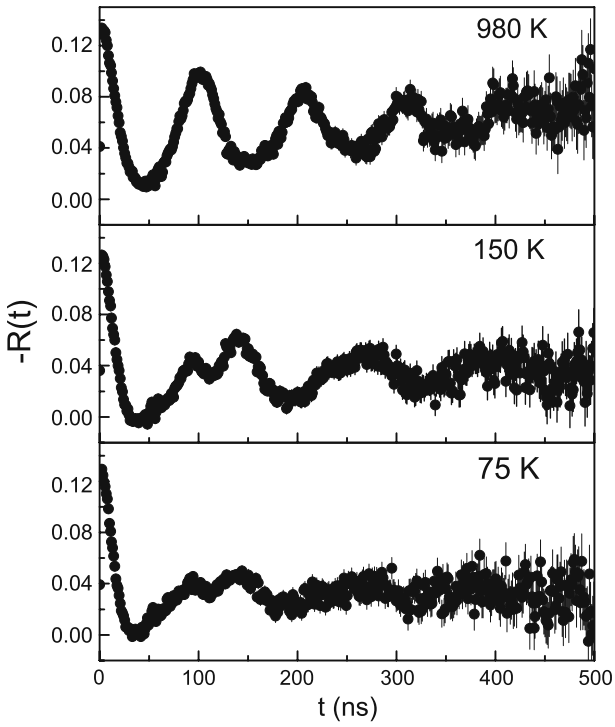

Fig. 1 PAC spectra measured with ^{111}Cd at indicated temperatures

and 98 K for ^{111}Cd and ^{140}Ce, respectively. We have no explanation for this difference yet. A similar behavior was observed in GdNiIn in a previous work and the possibility that this difference is sample dependent was ruled out because exactly the same transition temperature was observed in three different GdNiIn samples measured with ^{111}Cd probes [7]. The saturation value of B_{hf} for ^{111}Cd (10.50 T) is much larger than the value for ^{140}Ce (6.45 T). This observation can be ascribed to the difference in the bond distances between the probes to the magnetic Gd ion (∼3.2 Å for ^{111}Cd-Gd and ∼3.9 Å for ^{140}Ce-Gd) as well as to the polarization of the inner shells in the ^{140}Ce probe atom due to a spin polarization of the 5d electrons. This core polarization gives origin to a negative contribution to B_{hf}.

Measurements of quadrupole interaction with ^{111}Cd probe were performed above T_C up to 980 K. The results show that the fraction of probe nuclei occupying In sites increases with temperature and reaches around 95% above 500 K. If we admit the possibility that probes occupy both Gd and In sites, this increase can be interpreted as the existence of disorder in the In atoms occupying Gd sites, which is almost completely removed at high temperatures. A similar behavior was previously observed in the intermetallic compounds containing Pd and In [8]. Other explanation would be that if probes are trapped in defects as temperature increases, the defects are removed and the fraction of probes occupying In sites increases.

The saturation values of B_{hf} at ^{111}Cd for $GdNi_2$ [5], $GdAl_2$ [6], and Gd [9] and their correlation with respective Curie temperatures was made in reference 8. The data obtained for GdPdIn in the present work and that for GdNiIn [10] were added to this correlation as shown in Fig. 3 (top). A linear dependence of $B_{hf}(0)$ values

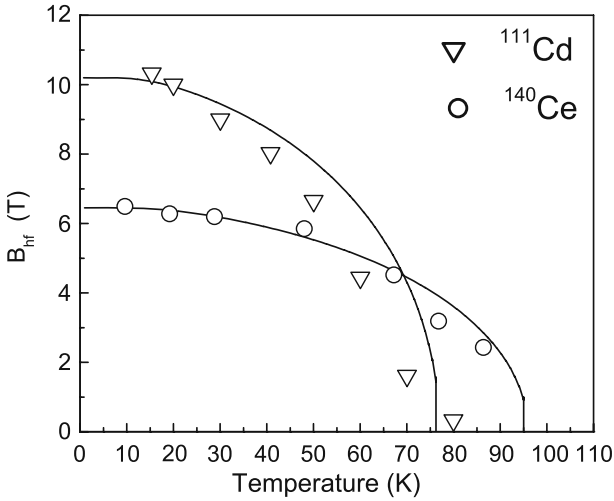

Fig. 2 Temperature dependence of the magnetic hyperfine field at In (*open triangles*) and Gd (*open circles*) sites in GdPdIn for ^{111}Cd and ^{140}Ce probes, respectively. The *solid lines* represents a Brillouin curve for J = 7/2

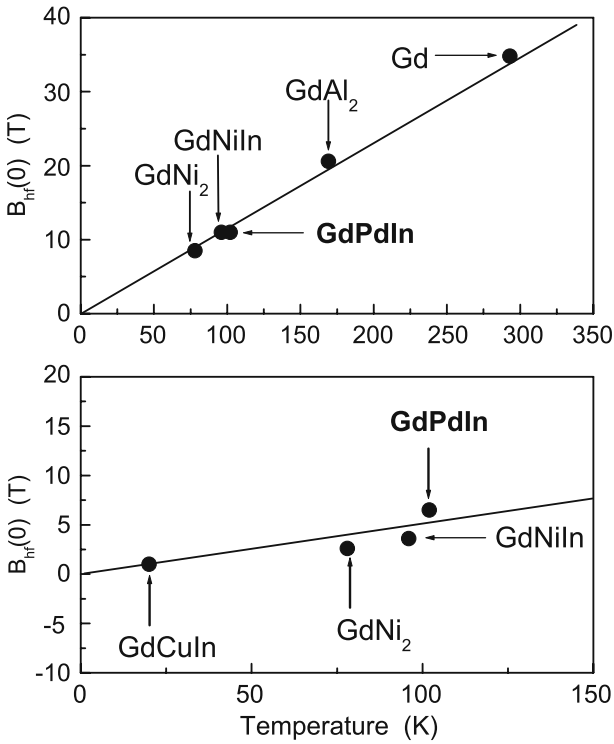

Fig. 3 The extrapolated magnetic hyperfine field $B_{hf}(0)$ at ^{111}Cd (*top*) and ^{140}Ce (*bottom*) in some Gd compounds as a function of the respective magnetic transition temperatures

with the T_C is clearly observed with $B_{hf}(0)/T_C = 0.12$ T/K. This value agrees with the value of 0.116 T/K reported in reference 8. The saturation values of $B_{hf}(0)$ at ^{140}Ce in GdPdIn, GdNiIn[10], $GdNi_2$ [11], and GdCuIn [11] are compared to the respective magnetic transition temperature of each host in Fig. 3 (bottom). One can observe

that to a good approximation $B_{hf}(0)$ is a linear function of the transition temperature here also. 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 $B_{hf}(0)$ at ^{140}Ce and the magnetic transition temperature shown in Fig. 3 (bottom) 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 for all the Gd compounds compared. Therefore, the ^{140}Ce probes in these cases behave 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.

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References

1. Balanda, M., Szytula, A., Guillot, M.: Magnetic properties of RPdIn (R=Gd-Er) compounds. J. Magn. Magn. Mater. **247**, 345–354 (2002)
2. Carbonari, A.W., Saxena, R.N., Pendl Jr., W., Mestnik-Filho, J., Attili, R.N., Olzon-Dionysio, M., Souza, S.D.: Magnetic hyperfine field in the Heusler alloys Co_2YZ Y = V, Nb, Ta, Cr; Z = Al, Ga. J. Magn. Magn. Mater. **163**, 313–321 (1996)
3. Dogra, R., Junqueira, A.C., Saxena, R.N., Carbonari, A.W., Mestnik-Filho, J., Morales, M.: Hyperfine interaction measurements in $LaCrO_3$ and $LaFeO_3$ perovskites using perturbed angular correlation spectroscopy. Phys. Rev. B **63**, 2241041–2241049 (2001)
4. Mestnik-Filho, J.: (Private communication)
5. Müller, S., de La Presa, P., Forker, M.: Perturbed angular correlation study of magnetic and eletric hyperfine interactions at ^{111}Cd in GdN_2 and $SmNi_2$. Hyperfine Interact. **133**, 59–64 (2001)
6. Forker, M., de La Presa, P., Olzon-Dionysio, M., de Souza, S.D.: Perturbed angular correlation study of magnetic and eletric hyperfine interactions at ^{111}Cd in $GdAl_2$. J. Magn. Magn. Mater. **226**, 1156–1158 (2001)
7. Lapolli, A.L., Saxena, R.N., Mestnik-Filho, J., Leite, D.M.T., Carbonari, A.W.: Local investigation of magnetism at R and In sites in RNiIn (R=Gd, Tb, Dy, Ho) compounds. J. Appl. Phys. **101**, 09D510-1–09D510-3 (2007)
8. Carbonari, A.W., Saxena, R.N., Mestnik-Filho, J., Cabrera-Pasca, G.A., Rao, M.N., Oliveira, J.R.B., Rizzuto, M.A.: Study of the effect of disorder on the local magnetism in Heusler alloys. J. Appl. Phys. **99**, 08J104–08J108 (2006)
9. Boström, L., Liljegren, G., Jonsson, B., Karlsson, E.: Hyperfine interaction for a cadmium impurity in gadolinium metal. Phys. Scripta **3**, 175–181 (1971)
10. Lapolli, A.L., Carbonari, A.W., Saxena, R.N., Mestnik-Filho, J.: Investigation of hyperfine interactions in GdNiIn compound. Hyperfine Interact. **158**, 157–161 (2004)
11. Lapolli, A.L.: Estudo de Interações Hiperfinas em Compostos Intermetálicos Gd(Ni,Pd,Cu)In, Tb(Ni,Pd)In, Dy(Ni,Pd)In e Ho(Ni,Pd)In. Dissertation, IPEN, São Paulo (2006)