

Hyperfine interactions at R and In sites in RNiIn (R = Gd, Tb, Dy, Ho) compounds measured by perturbed angular correlation spectroscopy

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Abstract Perturbed gamma–gamma angular correlation (PAC) technique was used to measure the magnetic hyperfine field (mhf) in RNiIn (R = Gd, Dy, Tb, Ho) intermetallic compounds using the $^{111}\text{In} \rightarrow ^{111}\text{Cd}$ and $^{140}\text{La} \rightarrow ^{140}\text{Ce}$ probe nuclei. The PAC spectra for ^{111}Cd measured above magnetic transition temperature show a major fraction with a well defined quadrupole interaction for all compounds except GdNiIn where a single frequency was observed. PAC measurements below T_C showed a combined electric quadrupole plus magnetic dipole interaction for ^{111}Cd probe at In sites, and a pure magnetic interaction for ^{140}Ce at R sites. The temperature dependence of mhf measured with ^{140}Ce at R sites shows that the values of fields drop to zero at temperatures around the expected T_C for each compound. However, in the measurements with ^{111}Cd at In sites, the mhf values become zero at temperatures which are smaller than T_C . The difference between the temperatures at which mhf is zero for ^{140}Ce and ^{111}Cd probes correlates with T_C . For each compound this difference decreases with T_C . The results are discussed in terms of the RKKY model for magnetic interactions and the existence of two magnetic systems, with distinct exchange interaction energies due to different types of atomic layers in these compounds.

Keywords Rare-earth compounds · PAC spectroscopy · Ferromagnetism · Magnetic hyperfine field

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1 Introduction

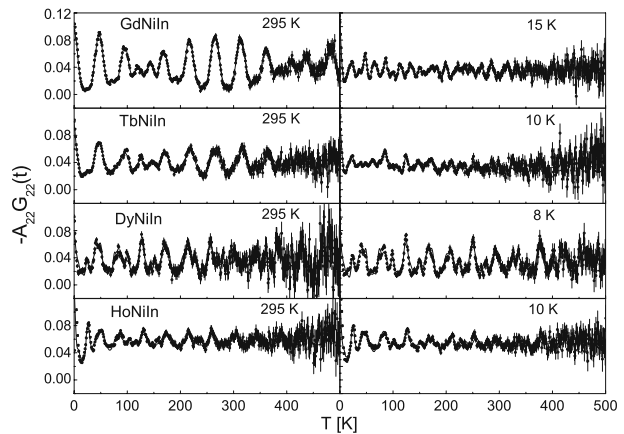
The investigation of hyperfine interactions in intermetallic magnetic compounds has been an important tool to better understand the origin of magnetic properties of these materials. In particular, due to very large thermal neutron absorption cross sections, little information is available from neutron diffraction studies about the details of the magnetic structure in some ternary intermetallic compounds containing rare-earth elements. Nevertheless, the investigation of the origin of the magnetic interactions on atomic scale is essential to better understand a wide variety of interesting magnetic properties in many families of rare-earth ternary compounds. The RNiIn family is formed by a series of compounds that crystallize in the ZrNiAl-prototype structure formed by magnetic R-Ni layers alternated with non-magnetic Ni-In layers. The magnetic R atoms form a triangular structure, which is a deformed Kagomé lattice. These compounds have been very little studied so far. In this family, GdNiIn, TbNiIn, DyNiIn and HoNiIn order ferromagnetically below 96, 70, 30 and 20 K, respectively [1]. Perturbed gamma–gamma angular correlation (PAC) technique has been used to investigate the temperature dependence of (efg) and magnetic hyperfine field (mhf) at In sites with ^{111}In - ^{111}Cd probe nuclei as well as (mhf) on rare earth atom, with ^{140}La - ^{140}Ce in RNiIn (R = Gd, Tb, Dy, Ho) compounds. Some of the results for the temperature dependence of mhf have been previously reported [2] and show that mhf measured with ^{140}Ce at R sites drop to zero at temperatures around the expected T_C for each compound but for the measurements with ^{111}Cd at In sites, the mhf values go to zero at smaller temperatures. In the present work, results of efg and mhf measurements are discussed.

2 Experimental procedure

Samples of RNiIn compound were prepared by repeatedly melting the constituent elements in an arc furnace under pure argon atmosphere. Carrier free ^{111}In nuclei were introduced into the sample by thermal diffusion. A separate sample was prepared for each compound with radioactive ^{140}La nuclei, produced by neutron irradiation of lanthanum metal in IEA-R1 reactor at IPEN, substituting about 0.1 % of R atoms. All the samples were annealed in vacuum for 48 h at 800°C.

The perturbed gamma–gamma angular correlation (PAC) measurements were carried out using a spectrometer consisting of four conical BaF₂ detectors with a conventional fast-slow coincidence electronic set-up. The well known 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 2,083 keV ($T_{1/2} = 3.45$ ns) in ^{140}Ce was used to measure the magnetic hyperfine field (B_{hf}) at Ce. The samples were measured in the temperature range of 8–300 K using a closed-cycle helium cryogenic device. The time resolution of the system was about 0.6 ns for both gamma cascades. A detailed description of the PAC method as well as measurements and analysis of data can be found elsewhere [3, 4].

Fig. 1 PAC spectra measured with ^{111}Cd probes at indicated temperatures for RNiIn compounds. The solid lines are the least squares fit to the experimental data



3 Results and discussion

Some of the PAC spectra measured with ^{111}Cd are shown in Fig. 1. The results for temperatures above the magnetic transition temperature show a major fraction (62% to 81%) with well defined quadrupole interaction for all compounds except GdNiIn for which a single frequency was observed. The additional interaction observed for the other three compounds is due to probe nuclei occupying some other site, probably the rare-earth sites, or trapped in vacancies. Other possibility would be that probe nuclei occupy In sites in a different compound formed during arc melting of constituents. In TbNiIn, at room temperature for instance, 81% of ^{111}Cd probes show an interaction with $\nu_Q = 80.8(1)$ MHz, $\delta = 2.4(1)\%$, and $\eta = 0.81(1)$ while 19% are associated to $\nu_Q = 88.1(1)$ MHz, $\delta = 0$, and $\eta = 1$.

At low temperatures, each compound shows a combined magnetic dipole plus electric quadrupole interaction and the measured spectra are characterized by a major quadrupole frequency and a temperature dependent magnetic dipole interaction. Table 1 shows the experimental values for V_{zz} and η as well as the extrapolated value for $B_{hf}(0)$ and the magnetic ordering temperature determined from the fitting of a Brillouin function for J spin of the R element to the experimental data for each compound. In all cases, below approximately reported Curie temperature a unique magnetic interaction was observed for ^{140}Ce at rare earth atom sites of the samples. Due to very small value the quadrupole moment of the 2,083 keV 4^+ state of ^{140}Ce almost pure magnetic dipole interactions were observed in the ferromagnetic phase of the samples.

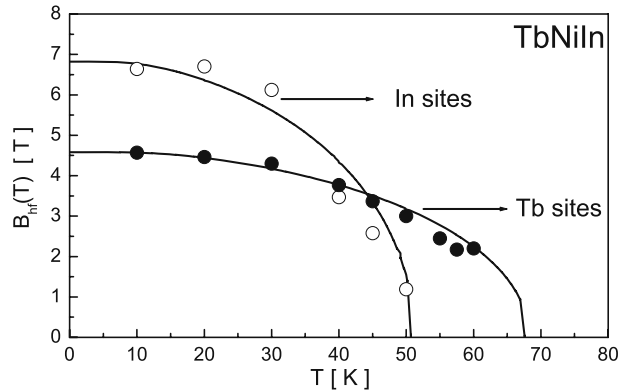
The results for the temperature dependence of the B_{hf} for all compounds, measured with ^{140}Ce and ^{111}Cd , show a significant difference in the temperatures below which the magnetic order starts (see Table 1). The possibility that this difference is sample dependent was ruled out mainly because the minor fraction observed in TbNiIn, which was assigned to ^{111}Cd probes occupying Tb sites, showed a magnetic interaction with $T_C = 68.3(1.2)$ K, very close to the reported value of 70 K for the Curie temperature, whereas for probes occupying In sites in the same sample

Table 1 Experimental values for some hyperfine parameters and magnetic transition temperatures for the studied compounds

Compound	${}^aV_{zz}$ (10^{21} V/m 2)	${}^a\eta$	$B_{hf}^{Cd}(0)$ (Tesla)	$B_{hf}^{Ce}(0)$ (Tesla)	T_{Cd} (K)	T_{Ce} (K)	T_C (K)
GdNiIn	4.10(4)	0.79(1)	11.9(2)	3.52(7)	56.4(5)	95.8(7)	96
TbNiIn	4.03(4)	0.80(1)	6.8(1)	3.02(8)	51.2(4)	71.3(9)	70
DyNiIn	4.03(4)	0.80(2)	2.7(1)	2.05(7)	26.3(3)	29.8(5)	30
HoNiIn	4.31(5)	1.00(3)	1.8(1)	1.72(7)	18.1(3)	20.2(5)	20

^ameasured at room temperature

Fig. 2 Temperature dependence of B_{hf} at Tb (full circles) and In (open circles) sites measured with ${}^{111}\text{Cd}$ in TbNiIn compound. The solid lines are the Brillouin curve for $S = 3$

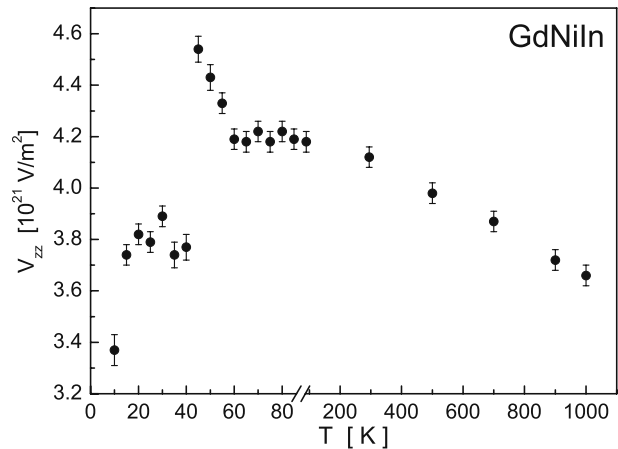


$T_{Cd} = 51.2(4)$ K (see Fig. 2). A strong evidence that other phases are not responsible for the observed difference in the magnetic ordering temperature is the fact that the GdNiIn compound for which this difference is larger, the same behavior was observed in three different GdNiIn samples measured with ${}^{111}\text{Cd}$ probes and only one single frequency was observed for each sample.

The extrapolated value $B_{hf}(0)$ at ${}^{111}\text{Cd}$ sites in GdNiIn was compared to the Curie temperature [5] and the result showed a ratio of $B_{hf}/T_C = 0.12$ T/K that agrees very well with the value $B_{hf}/T_C = 0.116$ T/K previously reported by Forker et al. [6] where they compared the saturation value of B_{hf} at ${}^{111}\text{Cd}$ sites in some Gd compounds. The saturation value of B_{hf} at ${}^{140}\text{Ce}$ on Gd sites in GdNiIn was also compared to the respective transition temperature along with other Gd compounds and showed a linear behavior as well [7]. The linear relation between B_{hf} at ${}^{140}\text{Ce}$ and the magnetic transition temperature may imply that the main contribution to the B_{hf} comes from the CEP at the probe site via Fermi contact field from s-electrons. The main contribution to the magnetic hyperfine field at rare-earth nuclei is supposed to come from the orbital angular momentum of 4f-electrons with fields of the order of hundreds of Tesla. In free Ce^{3+} ion, $B_{hf} \sim 185$ T [8] and, as the $B_{hf}(0)$ for ${}^{140}\text{Ce}$ are much smaller than this value for all studied compounds, we conclude that the probe in this case behaves as closed shell nuclei like ${}^{111}\text{Cd}$.

Figure 3 shows the temperature dependence of V_{zz} for GdNiIn. It can be seen that V_{zz} decreases linearly with temperature above 80 K. At lower temperatures however, the value increases when temperature decreases from 60 to 50 K, and then there is a sharp decrease at still lower temperatures. The angle between V_{zz} and B_{hf} at low

Fig. 3 Temperature dependence of V_{zz} at In sites measured with ^{111}Cd in GdNiIn compound



temperatures is around 30° and increases to around 60° at 50 K. We believe that such variation in the V_{zz} is related to the fact that the ordering temperature is different at ^{111}Cd sites. One possible explanation would be that B_{hf} changes its direction with temperature and, as the structure of the compounds is formed in layers, the intensity of the magnetism become higher in the plane of the rare-earth layer than in the perpendicular direction at this plane towards the layer that contains the In.

The difference in the temperatures at which B_{hf} goes to zero for ^{140}Ce probe at R sites and ^{111}Cd probe at In sites in each compound points to an evidence that the RNiIn is, in fact, a group of compounds, which present two distinct magnetic systems. One of these systems is the $(3\text{R}+\text{Ni})$ atomic layer at $z = 1/2$ within the $P62m$ space group whereas the other one is the $(3\text{In}+2\text{Ni})$ layer at $z = 0$. The exchange interaction that gives rise to magnetism in these compounds is larger within the atomic planes than the interaction between the planes. This explains the relative independence of the two magnetic systems. The measured MHF at the minor fraction of Cd probe atoms in TbNiIn exhibit a transition temperature similar to that obtained with Ce probes. This is an evidence that in this case the ^{111}Cd probes are also occupying the Ce sites which lie within the $(3\text{R}+\text{Ni})$ layers that present a higher magnetic transition temperature.

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