Hyperfine Interact
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Study of hyperfine interactions in the tetragonal $GdRh_2Si_2$ using PAC spectroscopy

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Abstract Hyperfine interactions were studied in the intermetallic compound $GdRh_2Si_2$ by perturbed angular correlation (PAC) technique using $^{181}Hf(^{181}Ta)$ probe nuclei. The measurements were performed in the temperature range 15–285 K. The PAC spectra above the antiferromagnetic ordering temperature of the $GdRh_2Si_2$ compound ($T_N \sim 106$ K), were analyzed using a model that included only electric quadrupole interactions. The observed major fraction was assigned to the $^{181}Hf(^{181}Ta)$ probe substituting the Gd atoms. The PAC spectra below Néel temperature were analyzed using combined electric quadrupole and magnetic dipole interactions. The B_{hf} value at Gd, measured at 15 K was found to be 1.4(1) T which, is smaller, when compared with the values obtained in this compound using other nuclear probes, ^{155}Gd ($B_{hf} \sim 30$ T) and ^{140}Ce ($B_{hf} \sim 26$ T). The present result using $^{181}Hf(^{181}Ta)$ probe is quite interesting since it shows that the contribution to B_{hf} at Gd due the host is smaller than other components which contribute to the hyperfine field. The temperature dependence of B_{hf} shows an anomalous behavior.

Keywords Magnetic hyperfine field · Rare earths compounds · PAC

1 Introduction

Intermetallic compounds, RRh_2Si_2 (R = rare earth), have been widely studied due to their many interesting physical properties such as long range magnetic order,

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superconductivity and Kondo effect among others [1–3]. These types of compounds crystallize in a $ThCr_2Si_2$ type structure, with *I4/mmm* space group. In particular the intermetallic compound $GdRh_2Si_2$ is characterized by a high Néel temperature ($T_N \sim 106$ K) in comparison with other members of the family [3]. The magnetic moment is localized only on the Gd atom, being oriented in the a-b plane. $GdRh_2Si_2$ has been studied in the past by Mössbauer Spectroscopy (MS) using ^{155}Gd [4, 5] and perturbed angular correlation (PAC) spectroscopy using highly diluted nuclear probe ^{140}Ce [6]. The results of magnetic hyperfine field (B_{hf}) measured with ^{140}Ce probe showed an anomalous temperature behavior. In the case of rare earths, the magnetic interactions are principally due to two contributions: the orbital contribution (B_{hf}^{orb}), as in the case of Ce atom, and the intense core polarization (B_{hf}^{cp}), in the case of the Gd atom. In many cases, these contributions are much larger than the host contribution also called transferred field. Therefore, one way to measure the transferred hyperfine field due to the host and its temperature dependence is to use a nuclear probe in which orbital and core polarization contributions are negligible. In this work we have studied the behavior of B_{hf} as a function of temperature using the highly diluted nuclear probe ^{181}Hf(^{181}Ta) in $GdRh_2Si_2$.

2 Experimental procedure

The samples of $GdRh_2Si_2$, were prepared by arc melting the stoichiometric quantities of metallic component with high purity (Gd = 99.9 %, Si = 99.9999 % and Rh = 99.99 %). The radioactive ^{181}Hf($t_{1/2} = 45$d) nuclear probe which, decays by β^- to the exited stated of ^{181}Ta, was introduced in the sample by adding a small quantity of hafnium metal (<0.1%), which had been previously irradiated with neutrons in the IEA-R1 research reactor at a flux of 3×10^{13}n/cm^2·s for 64 h. After melting, the sample was submitted to thermal treatment at 950 °C for 64 h. The PAC measurements were carried out with a four BaF_2 detector spectrometer with a slow-fast electronic set up for measuring delayed γ–γ coincidences. The PAC measurements were performed in a wide range of temperatures from 15 to 285 K. The $GdRh_2Si_2$ sample was examined by X-ray diffraction (XRD) in order to check crystal structure. The result showed that the sample had a single phase corresponding the tetragonal structure of $ThCr_2Si_2$ type, belonging to the *I4/mmm* space group.

3 Experimental result

The results of lattice parameters and unit cell volume of $GdRh_2Si_2$ determined from the analysis of XRD data are: a = 4.045 Å, b = 4.05 Å, c = 9.98 Å and vol = 163.293 Å^3. These results are in good agreement with those reported in the literature [5]. The PAC spectra were measured in the temperature range of 15–285 K. The spectra above transition temperature $T_N \sim 106$ K, were analyzed using a model including only electric quadrupolar interaction. As an example, PAC spectrum obtained at 285 K is shown in Fig. 1a. The parameter derived from fitting of the spectrum showed tree sites: a well-defined frequency $\nu_{Q1} = 325(2)$ MHz and $f_1 = 83$ %, and and highly distributed frequencies $\nu_{Q2} = 438(4)$ MHz and $f_2 = 11$ % and $\nu_{Q3} = 508(5)$ MHz and

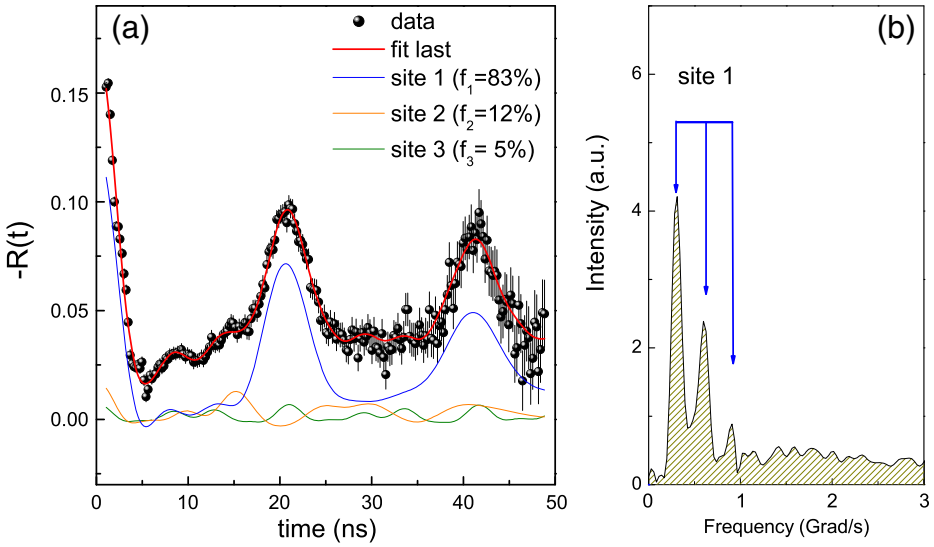

Fig. 1 **a** PAC spectrum at 285 K, the *solid lines* represent fitting of spectra with only electric quadrupole interactions for $GdRh_2Si_2$. **b** Fourier Transform Frequency majority, called a site 1

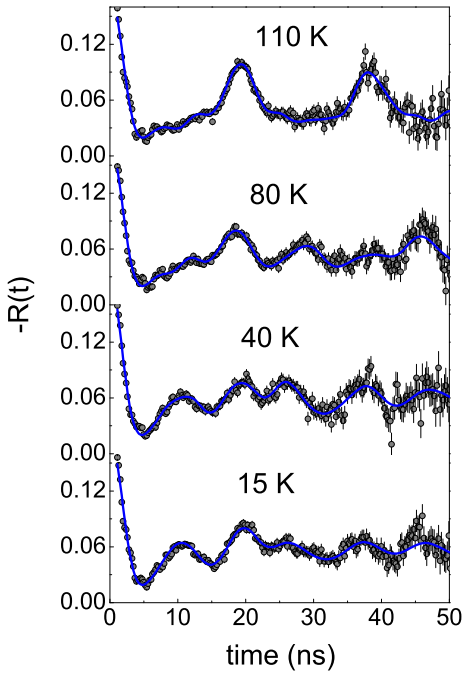

Fig. 2 The PAC perturbation functions for ^{181}Ta in $GdRh_2Si_2$ compounds at indicated temperatures. The *solid blue lines* are the least-squares fits of the theoretical function to the experimental data

$f_3 = 5$ %. The Fig. 1b shows the Fourier transform, in which one can observe the triplet corresponding to the majority fraction, called site 1. Below 200 K, the fraction f_3 disappeared completely while the fraction f_1 increased to about 90 %.

The PAC spectra, below the magnetic transition temperature, for $GdRh_2Si_2$, were analyzed using a model that included combined electric quadrupole and magnetic dipole interactions. Some of these PAC spectra are shown in Fig. 2 where the change in the modulation pattern due to magnetic interaction is clearly observed. Only a single well defined magnetic interaction was observed below the transition temperature. The origin of highly distributed minor fraction is not known at the moment. We shall focus only on the observed magnetic interaction and its temperature dependence in this work.

4 Discussion

In PAC experiments, when working with nuclear probe which is an impurity introduced externally in the system to be studied, an important issue is to determine the precise location of the nuclear probe within the crystal lattice. This information is especially very important when studying systems with different atoms, as is the case of $GdRh_2Si_2$ intermetallic compound studied with $^{181}Hf(^{181}Ta)$ probe. According to the crystal structure and symmetry considerations of this compound, the nuclear probe $^{181}Hf(^{181}Ta)$ could occupy any of the three possible atomic sites: gadolinium, rhodium or the silicon. We have however, assigned the major fraction observed in the PAC spectra as $^{181}Hf(^{181}Ta)$ replacing Gd atom based on the following observation: although there are no studies of neutron diffraction available for $GdRh_2Si_2$, it is known that compounds of the series RRh_2Si_2 have magnetic moment localized on rare earth atom only. For example in the study of ^{57}Fe-MS on RRh_2Si_2 realized by Anand et al. [7] no magnetic interaction was observed at the Rh site. Since in the present study of PAC on $GdRh_2Si_2$ using $^{181}Hf(^{181}Ta)$ probe we observe only one major site with magnetic interaction we believe that the probe is substituting the Gd site rather than Rh or Si sites.

According to the studies of ^{155}Gd Mössbauer spectroscopy in $GdRh_2Si_2$ at 4.2 K the value of B_{hf}, associated with the Gd atom was found to be 31.5 T [7, 8]. As mentioned earlier the contribution to B_{hf} in Gd will be due to polarization of the conduction electrons (B_{hf}^{cep}), due to host, and due to core polarization (B_{hf}^{cp}) from the spin ($S = 7/2$) of Gd atom. In this case the orbital contribution is insignificant (B_{hf}^{orb}) (angular momentum $\ell = 0$). According to the measurements with ^{155}Gd in $GdRh_2Si_2$ the B_{hf}^{cp} would be negative, $B_{hf}^{cp} = -34$ T, which is constant and does not depend on the system. Therefore $B_{hf}^{cep} \sim 2.5$ T for $GdRh_2Si_2$ [5]. The B_{hf} value obtained for ^{181}Ta at Gd measured at 15 K was ~1.45 T. This is smaller than the values obtained by ^{140}Ce-PAC measurement and ^{155}Gd-MS measurement. The result is quite interesting because it shows that the contribution of conduction electrons polarization in this system is small compared with the other components that contribute to the hyperfine field, namely orbital and core polarization observed using other nuclear probe such as ^{155}Gd and ^{140}Ce.

The temperature dependence of hyperfine field B_{hf} is shown in Fig. 3 along with Brillouin curve for $S = 7/2$ of Gd. The magnetic transition temperature $T_N \sim 106$ K determined from the present data is in good agreement with the literature value. The temperature dependence of the B_{hf} follows a behavior different from the Brillouin-type for $S = 7/2$ (Fig. 3). This difference may have its origin in the contribution due to the formation of local magnetic moments at impurity site due to open- d-orbital of Ta

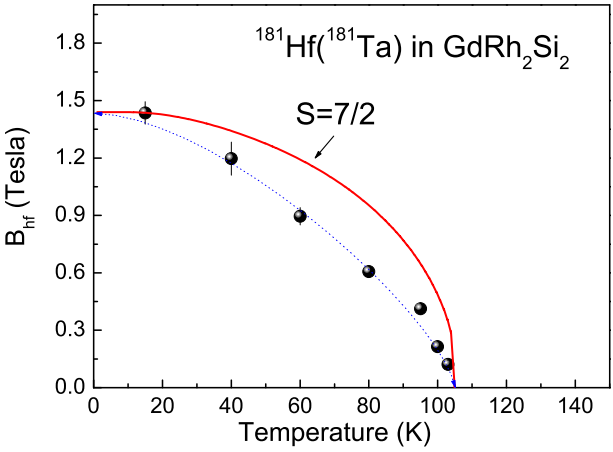

Fig. 3 **a** Temperature dependence of B_{hf} for the studied compounds. The *solid lines* represent the Brillouin curve for S = 7/2 of Gd, **b** Temperature dependence of Quadruple Frequency. *Dotted lines* are only to guide the eyes

atom, which would be sensitive to the temperature variation. A similar mechanism was observed for the ^{140}Ce nuclear probe in this compound [6]. The formation of localized states has been widely studied in the Laves phases RFe_2 with nuclear probe ^{181}Hf(^{181}Ta) [8]. We believe that the coupling between the open d-orbital of the Ta atom impurity and the d-band of the Rh atom is similar to the coupling between the 5d-band of Gd and d-band of Rh in $GdRh_2Si_2$ and $GdRh_2Ge_2$ described by Coehoorn at al. [9] and Mulder et al. [4].

5 Conclusion

New results of the temperature dependence of hyperfine fields measured with PAC spectroscopy, using the ^{181}Ta probe, for the intermetallic compound $GdRh_2Si_2$, are reported. The PAC results confirmed the magnetic transition temperature (∼106 K) reported previously in the literature. The results showed a small value of hyperfine field and anomalous temperature behavior of the B_{hf}. This behavior is attributed to hybridization of open d-band of Ta impurity with d-bands of the host, which are polarized by the magnetic ions, and responsible for the exchange interaction between the spins of the magnetic ions of the host and the Ta impurities.

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