2011 International Nuclear Atlantic Conference - INAC 2011
Belo Horizonte,MG, Brazil, October 24-28, 2011
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-04-5**

MAGNETIC HYPERFINE FIELD IN THE INTERMETALLIC COMPOUNDS $PrRh_2Si_2$ AND $GdRh_2Si_2$ MEASURED WITH PAC TECHNIQUE USING ^{140}Ce AS PROBE NUCLEI

G.A. Cabrera-Pasca, B. Bosch-Santos, R.N. Saxena, A.W. Carbonari

Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
gpasca@gmail.com

ABSTRACT

Perturbed gamma-gamma angular correlation (PAC) spectrsocopy has been used to measure the magnetic hyperfine field (B_{hf}) at rare-earth sites in the antiferromagnetic isostructural compounds $PrRh_2Si_2$ and $GdRh_2Si_2$ using $^{140}La \rightarrow ^{140}Ce$ as probe nuclei. The PAC measurements were performed in the temperature range from 10 K to 70 K in the case of $PrRh_2Si_2$ and from 10 K to 105 K in the case of $GdRh_2Si_2$. While the temperature dependence of B_{hf} for $PrRh_2Si_2$ follows a standard behavior expected for a second order magnetic transition, the temperature dependence of B_{hf} in the case of $GdRh_2Si_2$ is anomalous. The values of hyperfine field (B_{hf}) at 10 K, determined from the present measurements are 129.5(5) T and 25.6(4) T respectively for $PrRh_2Si_2$ and $GdRh_2Si_2$. The large difference in the hyperfine field values is explained by the difference in the mechanism for the formation of magnetic moments in the two compounds. The Neél temperatures (T_N) were determined to be 70.6(3) K and 106 (2) K respectively for $PrRh_2Si_2$ and $GdRh_2Si_2$ in good agreement with values found in the literature.

1. INTRODUCTION

The investigation of the behavior of the Ce ions embedded in a magnetic environment allows us to study interesting physical phenomena and helps us to understand the condensed matter physics as well. In particular, such studies provide important information concerning the physics of strongly correlated systems. These phenomena are attributed to the behavior of the 4f electron of cerium, which density of energy is located near the Fermi level when compared with other lanthanides. Therefore, the Ce ion is very sensitive to the chemical environment, in particular, the magnetic environment. In this paper we study the behavior of the Ce using hyperfine interactions techniques. The hyperfine interactions result from the interaction between the nuclear moments with extra-nuclear electromagnetic fields, and as such they are important in the study the electronic properties of the materials. Among the hyperfine interaction techniques are Mössbauer Spectroscopy (MS), nuclear magnetic resonance (NMR) and Perturbed Angular spectroscopy (PAC). Only PAC technique used in the present work allows us to study the behavior of the Ce ion using the ^{140}La- ^{140}Ce probe nuclei. The radioactive parent nuclei ^{140}La were introduced in the magnetic compounds $PrRh_2Si_2$ and $GdRh_2Si_2$ as highly diluted impurities partially substituting Pr and Gd atoms.

The compounds $PrRh_2Si_2$ and $GdRh_2Si_2$ crystallize in the prototype structure of $ThCr_2Si_2$ with *I4/mmm* space group. The magnetic properties of $PrRh_2Si_2$ have been studied by neutron diffraction, magnetization and resistivity measurements on polycrystalline samples [1]. These studies show an antiferromagnetic ordering for the compound with Neél temperature T_N

~70K. On the other hand, $GdRh_2Si_2$compound is characterized by T_N~106 K, which decreases when an external pressure is applied [2]. Earlier studies have shown that in $PrRh_2Si_2$ the spin moments are aligned in the c-axis due to the high uniaxial anisotropy [3] whereas the spins moments of $GdRh_2Si_2$ are parallel to the ab-plane [4].

2. EXPERIMENTAL

The samples of $PrRh_2Si_2$ and $GdRh_2Si_2$ were prepared by arc melting the constituent metals Gd(Pr) (99.9%), Rh (99.98%) and Si (99.9999%) in stoichiometric amounts, in the argon atmosphere purified with hot titanium getter followed by annealing in vacuum at 900°C during 5 days. The samples were analyzed by X-ray diffraction. The radioactive parent nuclei ^{140}La, obtained by the neutron irradiation of metallic La in the IEA-R1 reactor of IPEN, were introduced as highly dilute impurities (<0.1%) into the compounds during arc melting process. PAC measurements were carried out with a conventional fast-slow coincidence set-up using four conical BaF_2 detectors at the Hyperfine Interaction Laboratory at IPEN. Details about the PAC technique and experimental methodology can be found in the ref.5. Since the intermediate level in ^{140}Ce at 2083 keV with $J^{\pi} = 4^{+}$ used in the PAC measurement has a very small quadrupole moment, only the magnetic hyperfine interaction is observed with this probe even in samples with non cubic crystalline structure as is the case of $PrRh_2Si_2$ and $GdRh_2Si_2$ compounds which have tetragonal crystal structure.

3. RESULT AND DISCUSSION

The results of the X-ray diffraction shown in Fig. 1 were analyzed by using the Rietveld method and they have confirmed the presence of single phase in both compounds corresponding to the $ThCr_2Si_2$ structure with lattice parameters of a = 4.0746 Å, c = 10,1457 Å for $PrRh_2Si_2$ Å and a = 4.0406 Å, c = 9,9800 Å for $GdRh_2Si_2$.

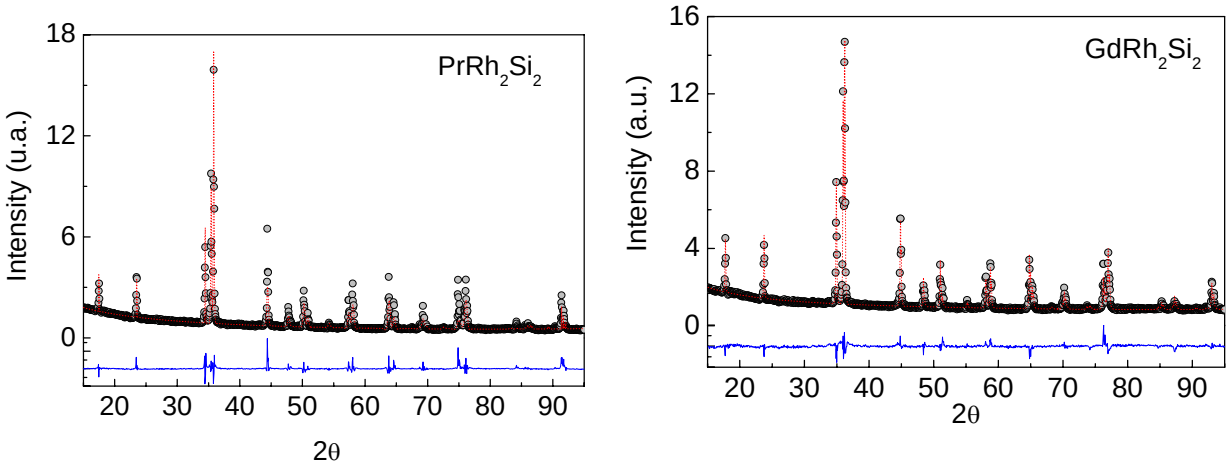

Figure 1: X-ray diffraction pattern for $PrRh_2Si_2$ and $GdRh_2Si_2$ samples. The solid line represents the calculated pattern with the Rietveld method. The residuals are shown in the lower part of each curve

PAC spectra measured below magnetic transition temperature were fitted using a model that included only magnetic dipole interactions, from which the Larmor frequency (ω_L) was determined. The experimental data were therefore least-squares fitted to the theoretical function $G_{22}(t) = 0.2[1 + 2\cos(2\omega_L t) + 2\cos(2\omega_L t)]$ [5]. Some of the measured PAC spectra are shown in Fig. 2.

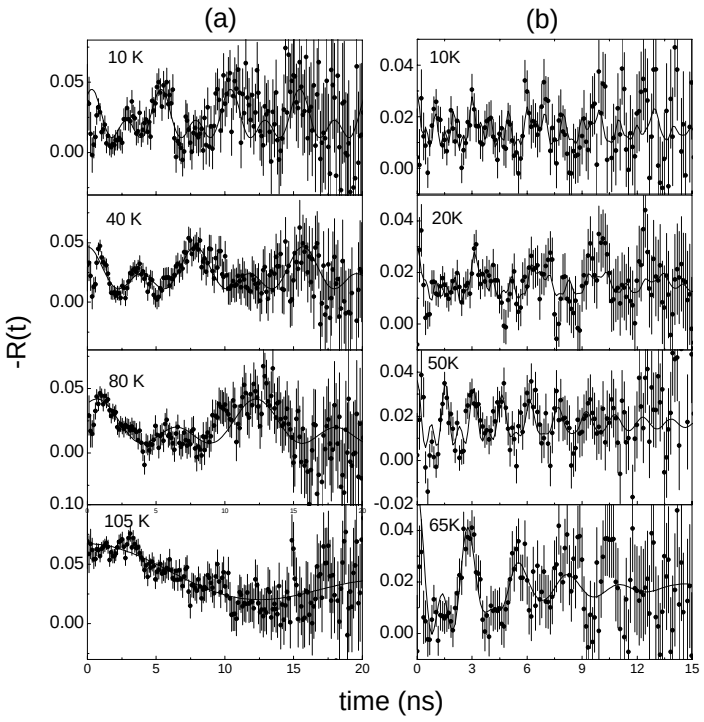

Figure 2: PAC spectra for studied intermetallic compounds (a) $GdRh_2Si_2$ and (b) $PrRh_2Si_2$ measured with ^{140}Ce at indicated temperatures. Solid lines are least-squares fits of the theoretical perturbation functions to the experimental data.

The temperature variation of the hyperfine field obtained from the PAC spectra for the samples are showed in Fig.3. The results permitted the determination of Neél temperatures for $PrRh_2Si_2$ and $GdRh_2Si_2$ as 70.6(3) K and 106(2) K respectively. These results are in agreement with the literature values.

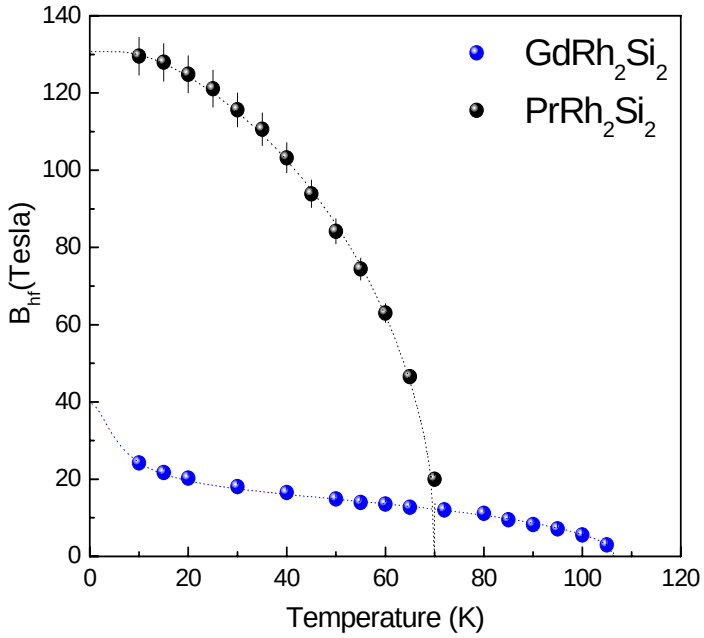

Figure 3: (a) Temperature dependence of the magnetic hyperfine field at ^{140}Ce probe in $GdRh_2Si_2$ and $PrRh_2Si_2$.

At 10 K (near magnetization saturation) a high value of B_{hf} = 129.5(5) T was observed at ^{140}Ce in $PrRh_2Si_2$, when compared to B_{hf} = 25.6(4) T for $GdRh_2Si_2$. This high value of B_{hf} suggests that the main contribution to B_{hf} in $PrRh_2Si_2$ comes from the orbital component of the Ce impurity, which would be larger than other components which contribute to the hyperfine field (dipolar field and Fermi contact field). However, this value is much smaller than that calculated for the free Ce ion (B_{hf} = 180(5) T). This difference is due to the crystalline field which directly affects the orbital component of B_{hf} [6]. The temperature dependence of B_{hf} at ^{140}Ce in $PrRh_2Si_2$ shows that the main component is polarized by the molecular field of the host and for this reason the behavior of hyperfine field follows the Brillouin function. On the other hand, the value of B_{hf} for $GdRh_2Si_2$ is found to be five times smaller than B_{hf} for $PrRh_2Si_2$, and shows anomalous temperature behavior. A similar behavior has been observed for Ce in metallic Gd. This behavior was studied by Leal and Troper [7] using an extension of the Daniel-Fridel model and the results of the performed calculations of B_{hf} at ^{140}Ce in Gd yielded the value of -53.7 T at T = 0 K. This value agrees very well with the value of 54 T measured by Thiel et al. [8] at 4.2 K. From these calculations the valence was estimated to be 3.25 instead of 3^+ for the Ce ions as impurity in Gd. As the B_{hf} is even smaller in $GdRh_2Si_2$ it is possible to suppose that the valence of Ce ions is greater than in the case of free ion Ce (3+).

4. CONCLUSIONS

The present results show that the investigation of the magnetic hyperfine behavior through PAC spectroscopy with ^{140}Ce is as probe nuclei is an important tool to characterize phase transitions in magnetic system containing rare earth atoms. In the present experiment antiferromagnetic transition temperatures were determined for $PrRh_2Si_2$ and in $GdRh_2Si_2$ that are in good agreement with the literature values. We conclude that the 4f electron of Ce is

well located at Ce probe ions in $PrRh_2Si_2$ but in the case of $GdRh_2Si_2$ Ce shows an intermediate valence.

ACKNOWLEDGMENTS

Partial support for this research was provided by the Fundacão de Amparo á Pesquisa do Estado de São Paulo (FAPESP). AWC and RNS thankfully acknowledge the support provided by CNPq in the form of research fellowships

REFERENCES

1. Z. Hossain; A.K. Rajarajan;V.K. Anand; C. Geibel and S.M. Yusuf, " Magnetic Propierties of $PrRh_2Si_2$ A Neutron Diffraction Study", Journal of Magnetism and Magnetic Materials, **v. 321**, p. 213-215, (2009).
2. M. Slaski; A. Szytula, "Crystal structure and Magnetic Properties of $REMe_2Si_2$ Compounds (RE= Gd,Dy,Ho, Er; Me=Ru,Rh,Pd,Ir)", Journal of the less-Common Metals, **v. 87**, p. L1-L3, (1982).
3. V.K. Anand;Z. Hossain; G. Behr; C. Chen, C. Geibel, "Magnetocrystalline Anisotropy and antiferomagnetic Phase Transition in $PrRh_2Si_2$",Jornal of Physics Condeses Matter, **v. 19**, p.506205 (7pp), (2007).
4. G. Gzjzek; V. Oestreich ; H. Schmidt; K. Latka; K. Tomala, "A study of compounds GdT_2Si_2 by Mössbauer spectroscopy and by bulk magnetization measurements", Journal of Magnetism and Magnetic Materials, **v. 79**, p. 42-56, (1989).
5. A.W. Carbonari; J. Mestnik-Filho; R.N. Saxena; V. Lalic, "Magnetic hyperfine interaction in $CeMn_2Ge_2$ and $CeMn_2Si_2$ measured by perturbed angular correlation spectroscopy", Phys. Rev. B **v.69**, 144425-1 144425-7 (2004).
6. K.R. Lea; M.J.M. Leask; W.P.J. Wolf., "The reasing of angular momentum degeneracy of f-electron terms by cubic crystal fields", Phys. Chem. Solids, **v.23**, p.1381-1405 (1962).
7. C.E. Leal; A.J. Troper, "Local magnetic moments and intermediate valence state of cerium impurities in ferromagnetic rare-earth metals", Appl. Phys., **v.75**, p.6767-6739 (1994).
8. T.A. Thiel; E. Gerdau; M. Bottcher. and G.Netz, "Temperature dependence of the hyperfine field of cerium in gadolinium", Hyperfine Interactions **v.9**, p.459 (1981)