Hyperfine interaction study of CeRh$_2$Si$_2$ with perturbed γ-γ angular correlation spectroscopy using ^{111}Cd and ^{140}Ce probes

G. A. Cabrera-Pasca,[a] A. W. Carbonari, R. N. Saxena, and B. Bosch-Santos
Instituto de Pesquisas Energéticas e Nucleares IPEN-CNEN/SP, São Paulo 05508-000, Brazil

The intermetallic compound CeRh$_2$Si$_2$ has been investigated by means of perturbed γ-γ angular correlation (PAC) spectroscopy using ^{111}In(^{111}Cd) and ^{140}La(^{140}Ce) probe nuclei. Measurements with ^{111}Cd were carried out in the temperature range of 8–295 K. The results showed a highly distributed and almost temperature independent quadrupole interaction. No magnetic interaction was observed with this probe down to the lowest measured temperature. Measurements with ^{140}La(^{140}Ce) probe were carried out in the temperature range of 8–36 K to determine the magnetic hyperfine field at Ce sites. A well defined magnetic interaction was observed at around 36 K. The PAC spectra below 25 K showed an additional magnetic interaction which was assigned to ^{140}Ce probe substituting Ce positions with a different magnetic structure. © *2010 American Institute of Physics.* [doi:10.1063/1.3367973]

I. INTRODUCTION

Strongly correlated electron systems (SCES) have introduced a new physics to the understanding of condensed matter in which phenomena such as heavy fermions, superconductivity, and a new series of magnetic behavior offer excellent opportunity for the study of these very interesting systems, especially those containing cerium as one of the component. Thus in the past few decades SCES have been studied using different experimental techniques such as neutron diffraction, magnetic susceptibility, and resistivity among several others[1,2] resulting in a large volume of information. However difficulties persist in understanding the behavior of $4f$ electrons of Ce in these systems. In this respect the study of hyperfine interaction at the Ce atom position in these systems should permit a better understanding of these systems. The objective of the present work is to use the perturbed γ-γ angular correlation (PAC) spectroscopy with ^{111}Cd and ^{140}Ce probes to obtain the hyperfine parameters in CeRh$_2$Si$_2$. The PAC spectroscopy is the only technique that uses ^{140}La(^{140}Ce) probe nuclei to determine the magnetic hyperfine field at Ce. It is well known that the compound CeRh$_2$Si$_2$ exists in two antiferromagnetic (AF) phases with the wave vectors $\mathbf{q}_1 = (1/2, 1/2, 0)$ below $T_{N1} = 36$ K and $\mathbf{q}_2 = (1/2, 1/2, 1/2)$ below $T_{N2} = 26$ K. The two phases coexist below 26 K.[1,2] The two AF phases are associated with magnetic moments of 1.86 μ_B and 1.69 μ_B, respectively, oriented along the c axis.[3]

II. EXPERIMENTAL PROCEDURE

The sample of CeRh$_2$Si$_2$ was prepared by arc melting the constituent metals Ce(99.9%), Rh(99.98%), and Si(99.9999%) in stoichiometric proportions followed by annealing in vacuum at 800 °C during five days. The sample was analyzed by x-ray diffraction, which confirmed a single phase corresponding to the ThCr$_2$Si$_2$ structure with $I4/mmm$ space group. PAC measurements were carried out with a four BaF$_2$ detector spectrometer using ^{140}La(^{140}Ce) and ^{111}In(^{111}Cd) nuclear probes. For the measurements with ^{111}In(^{111}Cd) nuclear probes a carrier free ^{111}In activity was thermally diffused in the sample at 850 °C for 12 h. ^{140}La was introduced in the sample by arc melting together the metallic components Ce, Rh, and Si with approximately 1% of La which was previously irradiated with neutrons in the IEA-R1 reactor at IPEN for 10 h at the flux of 3 $\times 10^{13}$ n cm^{-2} s^{-1} to produce ^{140}La. The arc melted alloy was then annealed in vacuum at 900 °C for three days. The gamma cascades of 171–245 keV in ^{111}Cd populated in the electron capture decay of ^{111}In and 329–487 keV in ^{140}Ce populated from the β^- decay of ^{140}La were used for the PAC measurements. The measurements with ^{111}Cd and ^{140}Ce probes were carried out in the temperature range of 8–295 K and 8–36 K, respectively, using a closed loop helium cryogenic system.

III. RESULTS AND DISCUSSION

While both electric and magnetic interactions may contribute in the case of measurements with ^{111}Cd, only magnetic interaction is expected for ^{140}Ce probe as the quadrupole moment of the intermediate state involved in the gamma cascade in this nucleus is known to be very small. The results of the measurements with ^{111}Cd below room temperature showed a highly distributed and almost temperature independent quadrupole interaction ($\delta \sim 0.5$). No magnetic interaction was observed. This result contrasts with the measurement in a similar compound CeMn$_2$Si$_2$ by Carbonari *et al.*,[4] where a well defined quadrupole frequency was observed. Comparison of the result with the first-principles calculations of the electric field gradient led these authors to conclude that the ^{111}In probe substitutes the Mn atom in CeMn$_2$Si$_2$. The measurements with ^{140}La(^{140}Ce) probe were carried out in the temperature range of 8–36 K, and some of the PAC

a)Electronic mail: gpasca@usp.br.

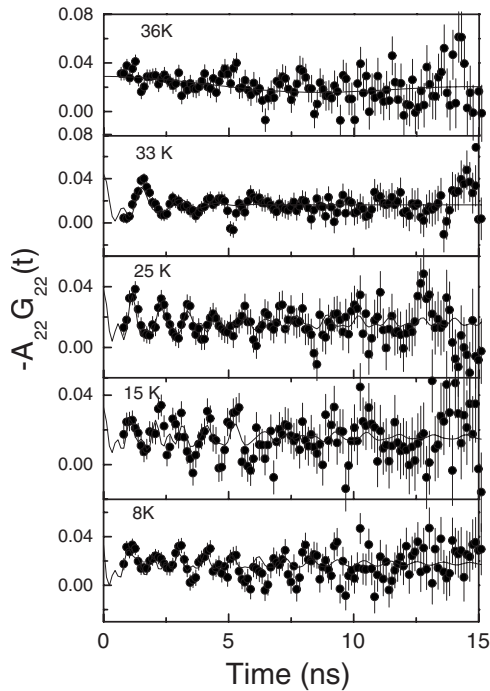FIG. 1. Perturbation functions for the ^{140}La(^{140}Ce) probe in CeRh$_2$Si$_2$ at some temperatures. The solid lines are the least square fit to the theoretical function.

FIG. 3. Temperature dependence of B_{hf} measured with ^{140}Ce probe. Full and open circles correspond to q_1 and q_2 magnetic structure, respectively. The continuous line represents the Brillouin function for $J=5/2$. Dotted lines are the guide to eyes.

spectra are shown in Fig. 1. The experimental data close to zero time were not considered for the fitting as they are affected by the prompt peak in the coincidence spectra.

A unique magnetic interaction was observed below 36 K. However, PAC spectra below 25 K showed an additional magnetic interaction. This observation is in complete agreement with the neutron diffraction study of CeRh$_2$Si$_2$ (Ref. 1) in which two distinct AF phases q_1 and q_2 were determined. In the temperature range of 36–25 K only q_1 phase exists while below 25 K both magnetic phases coexist. Relative fractions of the two phases determined from the present experiment as a function of temperature are shown in Fig. 2.

Temperature dependence of the hyperfine field (B_{hf}) for the two phases is shown in Fig. 3. The measured values of the hyperfine fields at 8 K are B_{hf} $(q_1)=130(1)$ T and $B_{hf}(q_2)=98(1)$ T, respectively. The temperature dependence of $B_{hf}(q_1)$ suggests a second order transition, nevertheless it
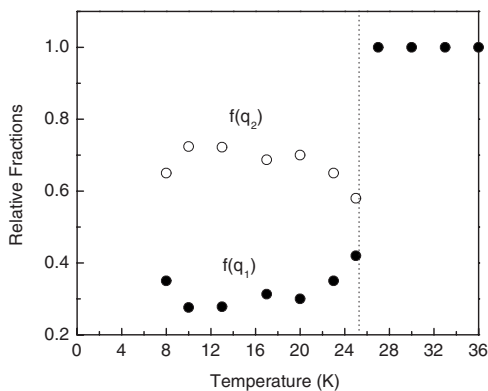

FIG. 2. Relative fractions of the magnetic phase q_1 (full circles) and q_2 (open circles) as a function of temperature.
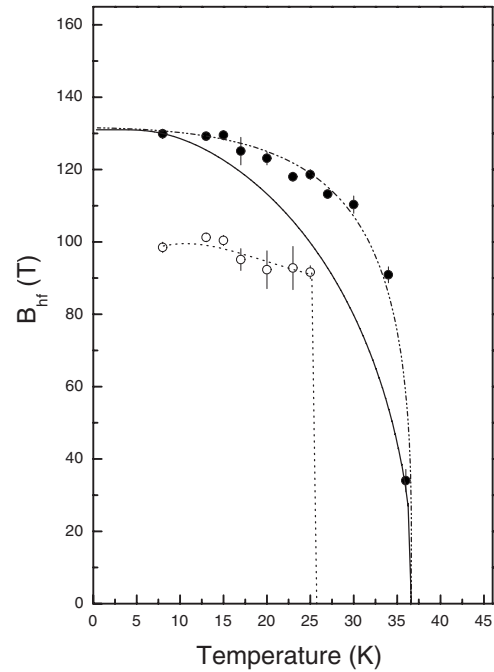
does not follow the Brillouin curve for $J=5/2$, shown as a continuous line in Fig. 3. The temperature dependence of $B_{hf}(q_2)$ is however significantly different. The field suddenly jumps from zero to a high value of approximately 90 T at around 25 K and then increases only slowly as temperature decreases. This may suggest a first order magnetic transition at about 25 K. Muon spin rotation (μSR) measurements on this compound by Yamamoto et al.[5] and NMR study by Kawasaki et al.[6] also suggest a first order transition for the q_2 magnetic phase. Kawasaki et al.[6] proposed that this magnetic structure is due to the independent AF domains. Present observation indicates that the fraction $f(q_2)$ gradually increases with the decrease in the temperature.

PAC spectroscopy measures only the effective B_{hf} at the probe nuclei, which, as is known to be due to three main contributions: orbital angular momentum, dipolar, and contact fields. Due to the presence of 4f electrons in Ce ion, it is expected that the major contribution comes from orbital angular momentum. This hypothesis was confirmed through the measurement of hyperfine field at Ce in CeIn$_3$ using ^{140}Ce probe nuclei and comparison with the first-principles calculation where individual contributions to B_{hf} can be determined explicitly.[7,8] Although the structures of CeIn$_3$ and CeRh$_2$Si$_2$ are different it may be worthwhile to compare the results for B_{hf} measured with ^{140}Ce in both compounds. CeIn$_3$ orders antiferromagnetically with localized magnetic moment of 0.65 μ_B at Ce ion, and shows a cubic structure where each Ce atom has six Ce atoms as 2nd nearest neighbor (2nn) ($d_{\text{Ce–Ce}} \sim 4.7$ Å). The measured B_{hf} at ^{140}Ce in the compound is 32.9(1) T.[7] On the other hand, the magnetic moment at Ce atoms in the q_1 magnetic structure in CeRh$_2$Si$_2$ is 1.86 μ_B with each Ce ion surrounded by 4 Ce atoms as 4nn ($d_{\text{Ce–Ce}} \sim 4.1$ Å). The measured B_{hf} value in

this case is 130(1) T which is roughly four times larger than for CeIn$_3$. This difference can be ascribed to the different crystal field effects in these compounds. The fundamental state of the Ce spin configuration in CeRh$_2$Si$_2$ is however not well known[9] and therefore it is not possible to make a more quantitative comparison with the configuration of CeIn$_3$.

IV. SUMMARY

PAC measurements of the hyperfine field (B_{hf}) at ^{140}Ce probes in CeRh$_2$Si$_2$ indicate the presence of two fractions with different B_{hf} values. These were attributed to probes in two AF structures q_1 and q_2, characterized by different ordering temperatures $T_{N1} \sim 36$ K and $T_{N2} \sim 25$ K, respectively. The magnetic transition at 25 K is characterized as a first order transition while the transition at 36 K is a second order transition. The fraction of the q_2 phase is found to increase with decreasing temperature. The value of B_{hf} at ^{140}Ce in CeRh$_2$Si$_2$ is considerably higher than corresponding value in CeIn$_3$ compound. This may be due to different crystal field effects in the two compounds.

[1] B. H. Grier, J. M. Lawrence, V. Murgai, and R. D. Parks, Phys. Rev. B **29**, 2664 (1984).

[2] T. Graf, M. F. Hundley, R. Modler, R. Movshovich, J. D. Thompson, D. Mandrus, R. A. Fisher, and N. E. Phillips, Phys. Rev. B **57**, 7442 (1998).

[3] S. Kawarazaki, M. Sato, Y. Miyako, N. Chigusa, K. Watanabe, and M. Nishi, Phys. Rev. B **61**, 4167 (2000).

[4] A. W. Carbonari, J. Mestnik-Filho, R. N. Saxena, and M. V. Lalic, Phys. Rev. B **69**, 144425 (2004).

[5] A. Yamamoto, S. Kawarazaki, Y. Miyako, K. Mishiyama, and K. Nagamine, Physica B **289–290**, 32 (2000).

[6] Y. Kawasaki, K. Ishida, Y. Kitaoka, and K. Asayama, Phys. Rev. B **58**, 8634 (1998).

[7] A. W. Carbonari, J. Mestnik-Filho, R. N. Saxena, and H. Saitovitch, Hyperfine Interact. **133**, 77 (2001).

[8] M. V. Lalić, J. Mestnik-Filho, A. W. Carbonari, R. N. Saxena, and H. Haas, Phys. Rev. B **65**, 054405 (2001).

[9] V. Vildosola, A. M. Llois, and M. Alouani, Phys. Rev. B **71**, 184420 (2005).