Hyperfine Interact (2007) 176:81–85
DOI 10.1007/s10751-008-9645-5

Study of hyperfine interactions in the intermetallic compound $CePd_2Si_2$ using PAC technique with ^{111}Cd as probe nuclei

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Published online: 6 August 2008

Abstract Perturbed $\gamma - \gamma$ angular correlation spectroscopy (PAC) has been used to investigate the hyperfine interactions in the intermetallic compound $CePd_2Si_2$ using $^{111}In \rightarrow ^{111}Cd$ probe nuclei. Samples of $CePd_2Si_2$ were prepared by melting constituent elements in an arc furnace under pure argon atmosphere. Carrier-free ^{111}In nuclei were introduced into the samples by thermal diffusion at 800°C in vacuum during 12 h. The measurements were performed in the temperature range of 4.2–300 K. Above the magnetic transition temperature ($T_N = 10$ K), the results show two distinct and well defined quadrupole interactions that were assigned to probe nuclei occupying Ce and Si sites in the compound. The quadrupole frequencies were found to decrease linearly with increasing temperature. The PAC spectra taken below 10 K were analyzed with a model including combined electric quadrupole plus magnetic dipole interactions, from which the hyperfine magnetic field was determined.

Keywords Magnetic hyperfine field · Ce compounds · PAC

PACS 75.30.Mb · 75.50.Ee · 76.80.+y

1 Introduction

Strongly correlated electrons systems (SCE) constitute some of the most fascinating and diverse research fields in physics. SCE generally originate from coulomb interaction. Although this interaction, is always present in charged particle systems, its effects are mostly pronounced in certain types of materials than in others. In some material classes, strong correlations lead to a completely new physics. Some of these types of materials are the intermetallic compounds based on Ce. One good example is

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the $CePd_2Si_2$ alloy that presents many fundamental state behaviors such as Kondo-type spin fluctuations, superconductivity at high pressure, antiferromagnetic phase limit and the non-magnetic phase and the long range magnetic order. It is believed that these behaviors occur due to the hybridization between conduction electrons and f-electrons (c–f hybridization). The competition between Ruderman–Kittel–Kasuya–Yosida (RKKY) and Kondo interaction, results from hybridization of c-f electrons. In the case of a weak hybridization, the RKKY interaction is dominant, that produces a long range magnetic order. In contrast, in the case of a strong hybridization, a Kondo interaction is dominant and produces valence fluctuations and non-magnetic state.

$CePd_2Si_2$ compound has the $ThCr_2Si_2$ crystal structure type that belongs to the *I4/mmm* space group. Cerium atoms form a body centered tetragonal (bct) simple sublattice. The $CePd_2Si_2$ compound has been studied in the past by different techniques, such as neutron scattering [1], which determined the magnetic structure below antiferromagnetic ordering temperature ($T_N = 10$K) and susceptibility measurements, which showed a large Curie–Weiss temperature of $\Theta \approx 60$ K and a small value for the magnetic moment ($0.62\mu_B$) [1–3]. The small value of the magnetic moment is probably due to the Kondo effect or the crystalline field effect.

Time differential perturbed $\gamma - \gamma$ angular correlation technique (TDPAC) was used to determine the hyperfine parameters, such as the electric field gradient (*efg*) and the magnetic hyperfine field (mhf) using ^{111}Cd as probe nuclei. This method offers high sensitivity to local distance variation in crystal lattice and, consequently can be used to follow changes such as magnetic ordering, bond distances, symmetry, defect trapping, etc., on microscopic scale by measuring the *efg* and mhf at probe sites.

2 Experiment

Samples of $CePd_2Si_2$ were prepared by repeated arc melting the pure metals (Ce 99.99%, Pd 99.9998% and Si 99.999%) in stoichiometric quantities in argon purified with a hot titanium getterer. Samples were annealed for 5 days at 700°C in vacuum. The radioactive ^{111}In was introduced in the samples through thermal diffusion at 800°C during 12 h in high vacuum.

PAC measurements were performed using a standard spectrometer with four BaF_2 detectors arranged in a planar 90–180° geometry, associated with an electronic system for delayed $\gamma - \gamma$ coincidence measurements. A detailed description of PAC method as well as the experimental procedure can be found elsewhere [4]. The measurements were performed in a temperature range of 8–300 K using a closed cycle helium cryogenic device, and with a liquid helium cryostat below 8 K.

3 Results and discussions

X-ray diffraction analysis of the sample confirmed a single phase tetragonal structure for the compound. Some of the PAC spectra and their respective Fourier transform, measured in the temperature range 4.2–300 K are shown in Fig. 1a. PAC spectra above the transition temperature ($T_N = 10$K) showed two well defined quadrupole

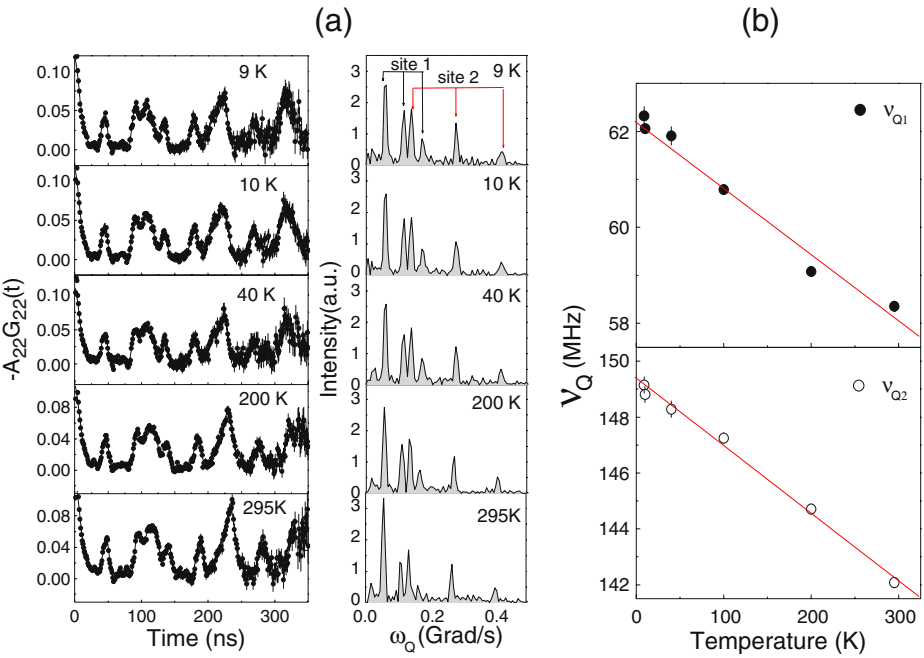

Fig. 1 **a** The perturbation functions and their Fourier transforms for ^{111}Cd probe in $CePd_2Si_2$ compound at various temperatures. *Solid lines* are the least-squares fits of the theoretical function to the experimental data. **b** Temperature dependence of the quadrupole interaction frequencies ν_Q for ^{111}Cd probe in $CePd_2Si_2$. *Solid lines* are the linear fit to the data

interactions with parameters: $\nu_{Q1} = 148.2(3)$ MHz f_1 60%, $\eta_1 = 0.1$ and $\nu_{Q2} = 61.9(3)$ MHz, f_2 40%, $\eta_2 = 0.05$ respectively obtained at 40 K. These interactions were associated to probe nuclei ^{111}In substituting Si and Ce atoms respectively. These assignments are consistent with the preliminary calculations performed within the framework of the density functional theory (DFT) in the WIEN2k computer code [5]. The results of the calculations of *efg* at Ce, Pd and Si in $CePd_2Si_2$ are (0.28, 0.22 and 1.32) $\times 10^{21}$Vm^{-2} respectively. As the calculations were made in pure compound and not including cd impurity at different atomic sites they should not be taken at their face value but only to indicate a trend in the calculated parameters. Calculated values of *efg* at Ce and Pd sites are much smaller than that for Si site. The calculations however do not rule out the possibility of the lower frequency being associated to Pd site. In an earlier PAC measurements on Heusler alloys Pd_2MnZ (Z=Ge, In, Sn, and Sb) no evidence was found that ^{111}In(^{111}Cd) probe substitutes Pd sites [6] most probably due to lack of chemical affinity between In and Pd. We therefore believe that the lower frequency corresponds to Ce site rather than Pd. The quadrupole frequencies decrease almost linearly with increasing temperature. The temperature dependence of the quadrupole frequencies are shown in Fig. 1b.

Below the magnetic transition temperature, PAC spectra were fitted using a model that included combined electric quadrupole plus a magnetic dipole interaction from which the Larmor frequency ω_L was determined (see Fig. 2a). Analysis of the spectra showed a single magnetic interaction which was associated to probe nuclei ^{111}In

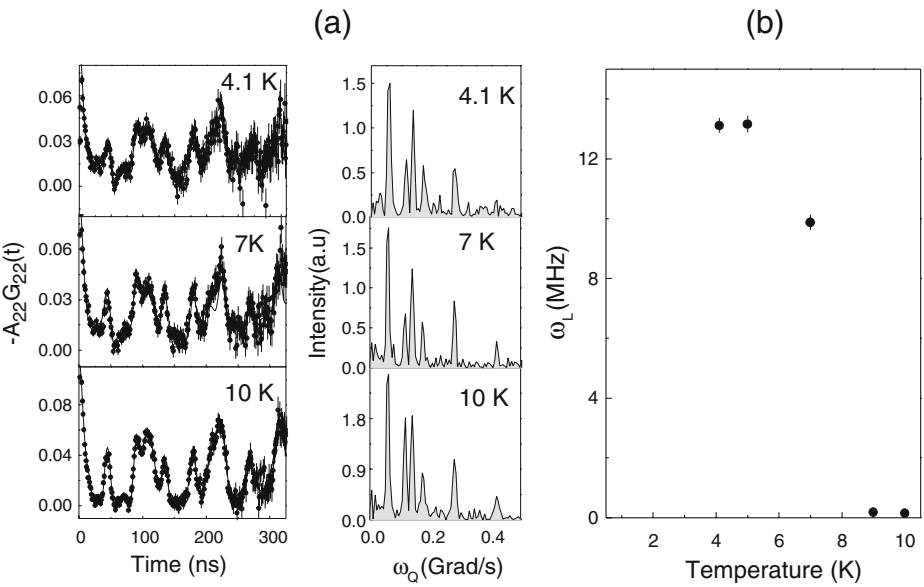

Fig. 2 **a** PAC spectra below magnetic transition temperature and respective Fourier Transforms. **b** Temperature dependence of the magnetic frequency

substituting Ce site. This assignment is once again based on the preliminary ab initio calculations that predict a relatively strong magnetic hyperfine field only on Ce in this compound. The calculated contact fields at Ce, Pd and Si are 13.6 T, 0.0 and 0.6 T respectively while the experimental value at 4.2 K is 5.6(2) T. This result is also an strong indication that $\nu_{Q2} = 61.9(3)$ MHz should be assigned to ^{111}Cd probes located at Ce sites and not at Pd sites. The temperature dependence of the Larmor frequency is presented in Fig. 2b. Although further measurements are needed at temperatures between 6-9 K there is a clear indication that the mhf has a normal Brillouin type behavior. Since only Ce has a magnetic moment in this compound, we deduce that the field at Ce originates from the 4f–5d interaction.

PAC technique has been shown to be efficient for measuring the hyperfine parameter of ^{111}Cd in $CePd_2Si_2$. Further measurements are in progress using ^{140}Ce probe nuclei to further elucidate the magnetic properties of $CePd_2Si_2$.

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

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