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Investigation of Hyperfine Interactions in $CeIn_3$ by TDPAC

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Abstract. Magnetic hyperfine fields (*mhf*) at ^{111}Cd and ^{140}Ce nuclei, dilutely substituting the In and Ce sites, respectively, have been measured in the intermetallic compound $CeIn_3$ using perturbed angular correlation technique. A pure electric quadrupole interaction with an axially symmetric electric field gradient was observed at ^{111}In(EC) ^{111}Cd probe nuclei at room temperature while a combined magnetic dipole and electric quadrupole interaction is observed below 10 K. Below the ordering temperature, only a magnetic interaction is observed at ^{140}La(β^-) ^{140}Ce probe. The values of *mhf* measured experimentally as a function of temperature are discussed in terms of critical behavior.

Key words: magnetic hyperfine field, heavy fermions, perturbed γ–γ angular correlation, critical exponent.

1. Introduction

The intermetallic compound $CeIn_3$ is a concentrated Kondo system with a heavy fermion behavior at low temperatures and exhibits an antiferromagnetic order below 10 K with saturated ordered moment of $0.65\mu_B$ per Ce atom [1]. There is a competition between the RKKY interaction and the Kondo effect in this compound. At low temperature the RKKY interaction predominates and the compound orders antiferromagnetically. Under increasing hydrostatic pressure, the Kondo state is stabilized and the antiferromagnetic state becomes unstable resulting in decreasing T_N values approaching to 0 K [2].

The $CeIn_3$ compound has a cubic structure of the $AuCu_3$ prototype with a lattice parameter of 4.63 Å, where Ce atoms are located at the corners and In atoms are located at the face-centered positions of a cubic unit cell. The In atoms are surrounded by Ce atoms and, due to noncubic symmetry, there is an electric field gradient (*efg*) at the In sites. In the present work, the Time Differential Perturbed γ–γ Angular Correlation (TDPAC) technique was used to measure the magnetic hyperfine field (*mhf*) at both Ce and In sites in $CeIn_3$.

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2. Experimental

The samples of $CeIn_3$ were prepared by arc melting the constituent elements (99.99%) under argon atmosphere along with the radioactive tracers (^{111}In and ^{140}La) followed by a 48 h annealing at 600°C in vacuum in each case. The crystal structure of the samples were checked by X-ray diffraction. The magnetic measurements were carried out in a SQUID magnetometer.

The TDPAC measurements in $CeIn_3$ samples were carried out with a conventional fast-slow coincidence set-up with four BaF_2 detectors. The γ-cascade of (172–245) keV, populated from the decay of ^{111}In with an intermediate nuclear level at 245 keV ($T_{1/2} = 84.5$ ns) in ^{111}Cd, was used for the measurement at the In site. The γ-cascade of (329–487) keV populated from the decay of ^{140}La with an intermediate nuclear level at 2083 keV ($T_{1/2} = 3.45$ ns) in ^{140}Ce was used to measure the *mhf* at the Ce site. The samples were measured in the temperature range of 4.2–295 K by using a closed cycle helium cryogenic device and a liquid helium cryostat. The time resolution of the system was about 0.8 ns for both γ-cascades.

The PAC method is based on the observation of the hyperfine interaction of nuclear moments with magnetic field or *efg*. A detailed description of this method can be found elsewhere [3, 4]. The perturbation factor $G_{22}(t)$ of the correlation function contains detailed information about the hyperfine interaction. The perturbation factor allows the determination of the Larmor frequency $\omega_L = \mu_N g H_{hf}/\hbar$ in the case of a magnetic dipole interaction, and the nuclear quadrupole frequency $\omega_Q = eQV_{zz}/4I(2I - 1)\hbar$ and the asymmetry parameter $\eta = (V_{xx} - V_{yy})/V_{zz}$, (where V_{xx}, V_{yy}, and V_{zz} are the components of the *efg* tensor in its principal axis system) in the case of a static quadrupole interaction.

The combined magnetic dipole and electric quadrupole interactions were analyzed by using appropriate expressions [5]. The eigenvalues of the Hamiltonian matrix depend on the ratio $y = \omega_L/\omega_Q$, and the angles α and β of the *mhf* direction in the *efg* coordinate system [5]. The experimental data for temperatures above T_N were analyzed for pure quadrupole interaction whereas those at $T < T_N$ were analyzed using combined interaction.

3. Results and discussion

Some of the TDPAC spectra and their respective fast Fourier transforms are shown in Figures 1 and 2 for ^{111}Cd and ^{140}Ce probe nuclei, respectively. The solid curves are the least-squares fits of the experimental data to the appropriate function in each case. The TDPAC spectra for ^{111}Cd at room temperature show a unique quadrupole interaction with a sharp frequency $\omega_Q = 12.3(1)$ Mrad/s and $\eta = 0$. Below 10 K, all the spectra for ^{111}Cd show combined magnetic dipole and electric quadrupole interaction. Each spectrum is characterized by a single quadrupole frequency with $\eta = 0$, and a temperature dependent *mhf*. The small value of the ratio $y \approx 0.23$ and the angle $\theta = 72.5°$ observed are in agreement with the expected effect on the ω_1 and ω_3 frequency components of the quadrupole interaction for a weak magnetic

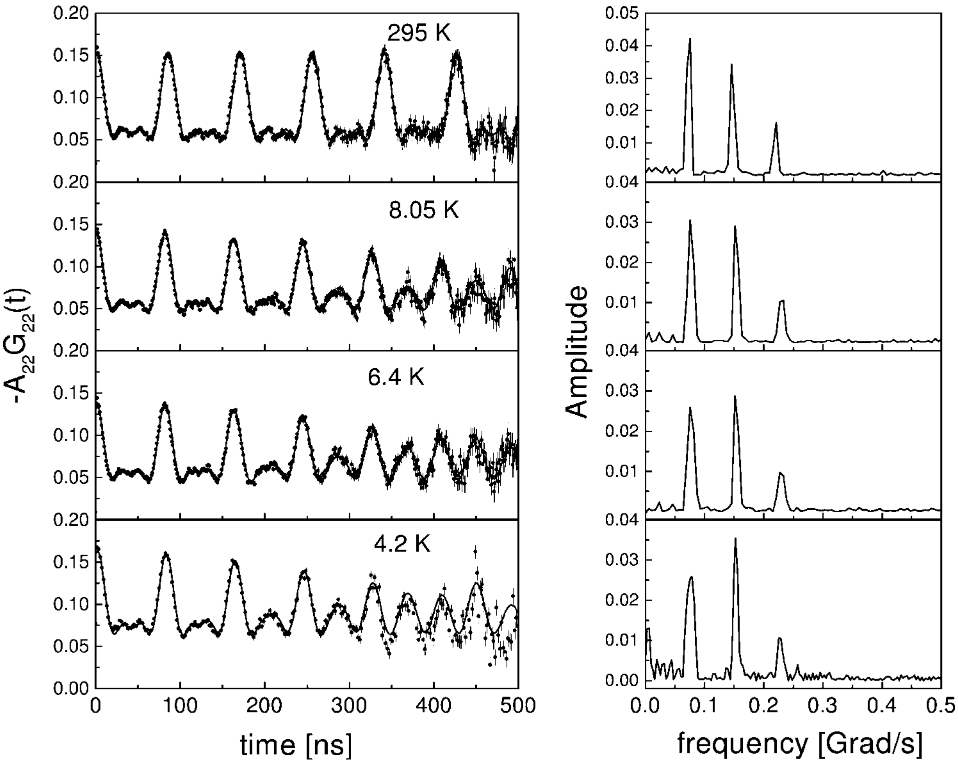

Figure 1. TDPAC and correspondent fast Fourier transform spectra for ^{111}Cd at In site in $CeIn_3$ compound measured at indicated temperatures.

hyperfine field ($y < 1$) nearly perpendicular to the principal axis of the *efg*. Under these conditions, the weak *mhf* causes only a small splitting of the component ω_1. The frequency component ω_2 is essentially unsplit and therefore the component $\omega_3 = \omega_1 + \omega_2$ also shows a small splitting (see [5] for details). As a consequence, the components ω_1 and ω_3 may be expected to show a broadening with increasing value of *mhf*. To some extent the Fourier transforms presented in Figure 1 show this effect. The amplitude of the components ω_1 and ω_3 is found to decrease with decreasing temperature as a result of the line broadening. As expected from the symmetry reasons the TDPAC spectrum for the ^{140}Ce probe at room temperature shows no quadrupole interaction. In the temperature range of 7–10 K, a pure magnetic interaction, ranging from 9 to 30 T, is observed with very small frequency distribution. The initial part of the spectra for ^{140}Ce probe was not included in the fitting because of the strong influence from the prompt coincidences.

The temperature dependence of *mhf* for ^{111}Cd at In site and for ^{140}Ce at Ce site are plotted in Figure 3. The solid curves are the least-square fits of the *mhf* values to the modified Curie–Weiss law $B(T) = B(0)(1 - T/T_N)^{\beta}$. The results of the fitting gave $\beta = 0.28(4)$, $B(0) = 0.24(1)$ T, $T_N = 9.7(2)$ K for ^{111}Cd probe,

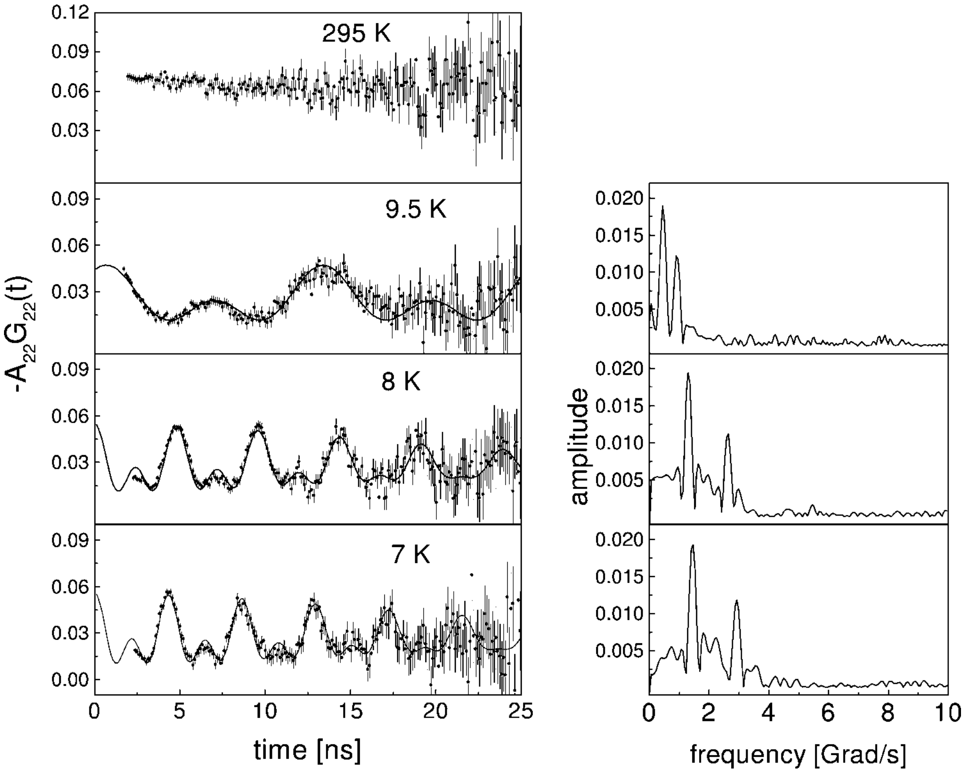

Figure 2. TDPAC and correspondent fast Fourier transform spectra for ^{140}Ce at In site in $CeIn_3$ compound measured at indicated temperatures.

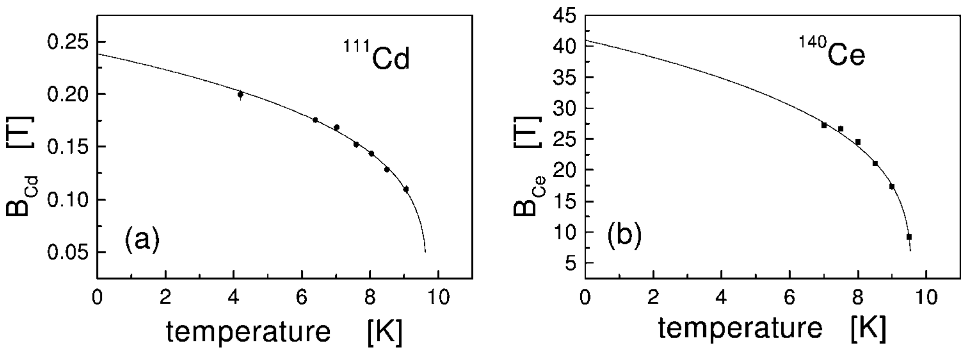

Figure 3. Temperature dependence of the magnetic hyperfine field (a) on ^{111}Cd at the In site, and (b) on ^{140}Ce at the Ce site for $CeIn_3$.

and $\beta = 0.30(3)$, $B(0) = 41(2)$ T, $T_N = 9.6(1)$ K for ^{140}Ce probe. The value of T_N is somewhat smaller compared to the value of 10.2 K measured by neutron diffraction [1] and the values for β are smaller than the theoretical value expected for a 3-dimensional isotropic Heisenberg magnet ($\beta \approx 0.38$).

4. Conclusions

In the present work, the application of a not very frequently used PAC probe, ^{140}La–^{140}Ce, has been successfully demonstrated for *mhf* measurements at the Ce site in the the $CeIn_3$ compound. At the In site of this compound, the results of the measurements in the temperature range of 4.2–10 K showed a combined interaction with an *efg* value $V_{ZZ} = 4.07 \times 10^{17}$ V/cm^2 and magnetic hyperfine field values varying from 0.1 to 0.2 T. In $CeIn_3$, the magnetic moments are aligned ferromagnetically in the (1 1 1) planes with adjacent planes antiparallel one to another [6]. As a result, the isotropic component of the magnetic hyperfine field at the In site is expected to vanish. The small *mhf* observed at ^{111}Cd on the In site was also observed by NQR technique on ^{115}In probe [7] and could originate from an anisotropic component of the magnetic hyperfine field. The higher values for the *mhf* at the ^{140}Ce site are mainly due to the orbital contribution of the 4f electrons. The critical exponents were determined for both sites.

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