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Hyperfine Interactions in CeT_2Ge_2 (T = Mn, Co) Heavy Fermions Compounds Measured by TDPAC

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Abstract. Time Differential Perturbed γ–γ Angular Correlation (TDPAC) technique was used to measure the magnetic hyperfine field at both Ge and Ce sites in $CeMn_2Ge_2$ and $CeCo_2Ge_2$ intermetallic compounds. The ^{111}In (^{111}Cd) probe nuclei was used to investigate the hyperfine interaction at Ge sites, while the ^{140}La (^{140}Ce) nuclei was used to measure the magnetic hyperfine field at Ce site. The present measurements cover the temperature ranges from 10–460 K for $CeMn_2Ge_2$ and 9–295 K for $CeCo_2Ge_2$, respectively. The result for ^{111}Cd probe showed two distinct electric quadrupole frequencies above magnetic transition temperatures, in both compounds and a combined interaction in the magnetic region. The temperature dependence of the magnetic hyperfine field at ^{111}Cd at Mn site for the $CeMn_2Ge_2$ compound showed a transition from ferromagnetic to antiferromagnetic phase around 320 K and from antiferromagnetic to paramagnetic phase at 420 K. While a small magnetic field was measured on ^{111}Cd at Co site, no magnetic field on ^{140}Ce site was observed in $CeCo_2Ge_2$.

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

1. Introduction

Highly correlated electron systems are usually called heavy Fermions and the term is generally used to designate rare earth and actinide metals, their alloys and intermetallic compounds. The exotic properties associated with these systems are related to a Fermi liquid behavior and, in a free electron model for a metal, refer to the conduction electron density of states at the Fermi level. $CeMn_2Ge_2$ and $CeCo_2Ge_2$ are ternary heavy Fermions compounds from the family RT_2X_2 (R = rare earth or actinide, T = transition metal and X = Si, Ge) with the body-centered tetragonal $ThCr_2Si_2$-type crystalline structure. The $CeMn_2X_2$ compounds are of particular interest because they are the only, in the whole family, which exhibit magnetic moments only at the Mn sites. In these compounds, the Mn atoms form a simple tetragonal lattice. It has been reported [1, 2] that the $CeMn_2Ge_2$ has a collinear commensurate antiferromagnetic ordering with $T_N = 415$ K. At 318 K, there is a transition to a incommensurate ferromagnetic component of the Mn moments in the c axis and a helical component in the ab plane. The $CeCo_2Ge_2$ has no magnetic mo-

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ment on the Co atoms and orders antiferromagnetically below 29 K. In the present work, the magnetic and quadrupole interactions at Ce, Mn and Co sites was investigated by time differential perturbed gamma–gamma angular correlation (TDPAC).

2. Experimental

The $CeMn_2Ge_2$ and $CeCo_2Ge_2$ samples were prepared by repeatedly melting the constituent elements (Ce 99.99%, Mn 99.9985%, Ge 99.9995%) along with the radioactive tracers under argon atmosphere purified with a hot titanium getterer. Radioactive probe nuclei used were carrier free ^{111}In and less than 0.1% of ^{140}La obtained by neutron irradiation of lanthanum metal. Samples were annealed under an atmosphere of ultrapure Ar for 48 h at 700°C. The structures of the samples were checked by X-ray diffraction and the magnetic characterization was carried out in a SQUID magnetometer.

The TDPAC measurements were carried out with a conventional fast-slow coincidence set-up with four conical BaF_2 detectors. The well known gamma cascade of 172–245 keV, populated from the decay of ^{111}In with an intermediate level with spin $I = 5/2^+$ at 245 keV ($T_{1/2} = 84.5$ ns) in ^{111}Cd, was used to investigate the hyperfine interaction in $CeMn_2Ge_2$ and $CeCo_2Ge_2$ samples. The gamma cascade of 329–487 keV populated from the decay of ^{140}La with an intermediate level with spin $I = 4^+$ at 2083 keV ($T_{1/2} = 3.45$ ns) in ^{140}Ce was used to measure the magnetic hyperfine field (*mhf*) at Ce. The samples were measured in the temperature range of 10–420 K by using a closed-cycle helium cryogenic device. The time resolution of the system was about 0.6 ns for both gamma cascades.

The TDPAC method is based on the observation of hyperfine interaction of nuclear moments with extra-nuclear magnetic field or electric field gradient (*efg*). A detailed description of the method can be found elsewhere [3, 4]. The perturbation factor $G_{22}(t)$ of the correlation function contains detailed information about the hyperfine interaction. Measurement of $G_{22}(t)$ allows the determination of the Larmor frequency $\omega_L = \mu_N g H_{\rm hf}/\hbar$, and the nuclear quadrupole frequency $\nu_Q = eQV_{zz}/h$ 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 from the known g-factor and quadrupole moment of the 245 keV state of ^{111}Cd and the g-factor of 2083 keV state of ^{140}Ce. The experimental data for temperatures above T_N were analyzed for static quadrupole interaction, while those at $T < T_N$ were analyzed for combined magnetic dipole and electric quadrupole interactions by using appropriate expressions [5].

3. Results and discussion

Some of the TDPAC spectra and their respective Fourier transforms for $CeMn_2Ge_2$ are shown in Figure 1 for ^{111}Cd probe nuclei. The solid curves are the least-squares fit of the experimental data to the appropriate function in each case. The

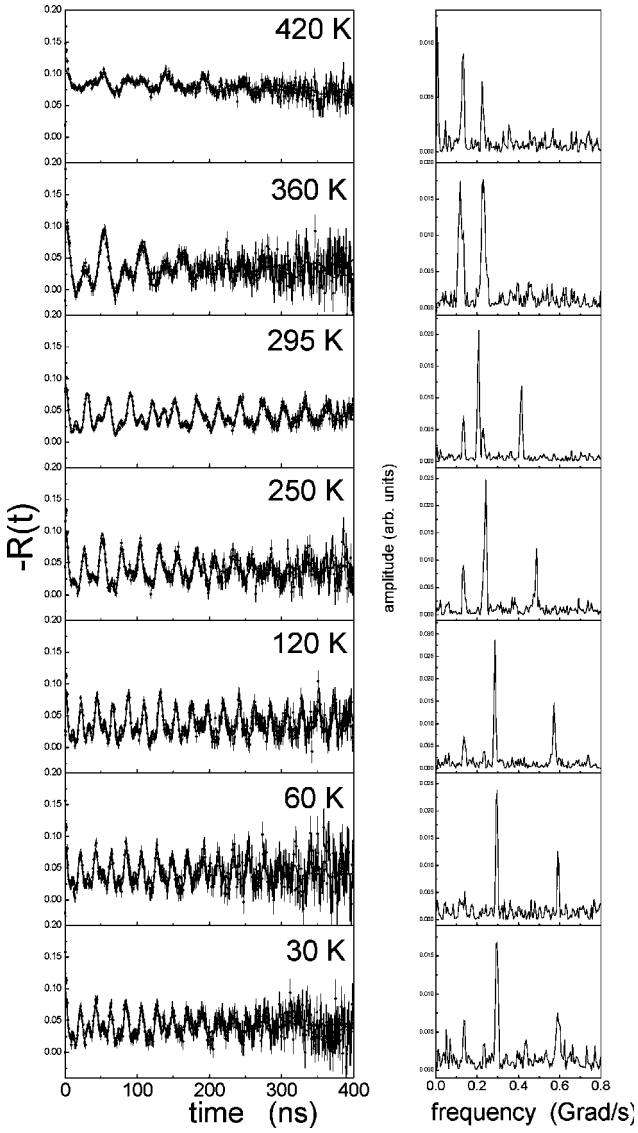

Figure 1. TDPAC and corresponding fast Fourier transform spectra for ^{111}Cd in $CeMn_2Ge_2$.

TDPAC spectra for ^{111}Cd at 420 K shows two quadrupole interactions with $\nu_{Q1} =$ 121.8(3) MHz, $\eta_1 = 0.3$, $\delta_1 \sim 0$ and $f_1 \sim 30\%$ and $\nu_{Q2} = 1.14(5)$ MHz, $\delta_2 \sim 0$, $\eta_2 = 0$ and $f_2 = 70\%$. By comparing these results to the values of *efg* at Ce, Mn and Ge sites obtained by preliminary ab initio calculations using full-potential linearized augmented plane waves FP-LAPW method [6], the lower frequency could be assigned to the Mn site. The higher frequency can be assigned either to the Ce or to the Ge site. All the spectra measured below 400 K, show combined magnetic dipole and electric quadrupole interactions. Each of these spectra are characterized

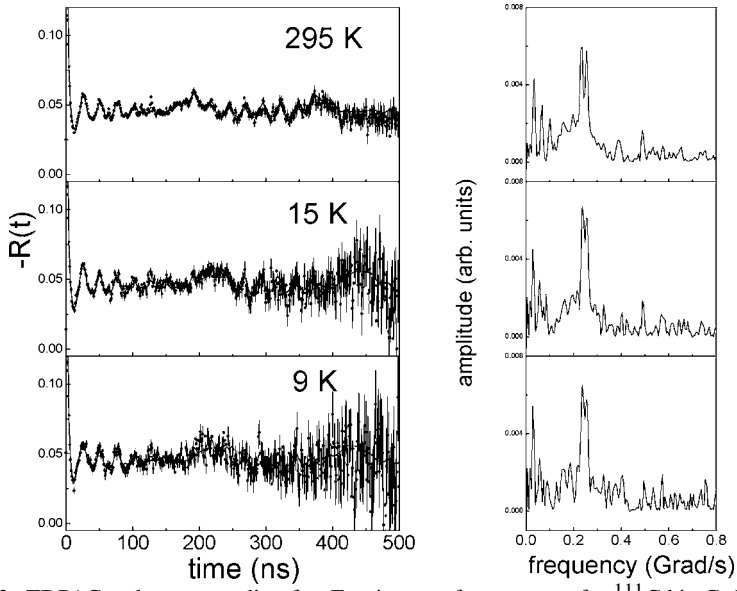

Figure 2. TDPAC and corresponding fast Fourier transform spectra for ^{111}Cd in $CeCo_2Ge_2$.

by two quadrupole frequencies, with approximately the same hyperfine parameters as obtained for 420 K, and two magnetic interactions. The magnetic interaction corresponding to the major fraction, which we assigned to the Mn site shows a strong temperature dependence with ω_L values ranging from ∼300 Mrad/s at 30 K to 47 Mrad/s at 400 K. The other frequency has a small value of ω_L, around 4 Mrad/s which does not appreciably change with temperature. While the TDPAC spectrum for ^{140}Ce probe measured above 320 K and 360 K did not show any interaction, at lower temperatures, a pure magnetic interaction is observed with very small frequency distribution ($\delta \approx 0.02$).

For $CeCo_2Ge_2$ samples, we obtained very similar TDPAC spectra in the entire temperature range of 15 K to 300 K, for both ^{111}Cd and ^{140}Ce probes. Some of the spectra measured with ^{111}Cd probe are shown in Figure 2. From the fitting of the spectra measured at 25 K and below, we obtained a small magnetic frequency (<1 Mrad/s) for the Co site. The effect of this small magnetic interaction can be better seen in the fast Fourier transform, as a very small perturbation in the amplitude of the electric quadrupole frequencies [7].

The temperature dependence of the Larmor frequency ω_L for ^{111}Cd at Mn site in $CeMn_2Ge_2$ is plotted in Figure 3. The behavior of the magnetic frequency for ^{111}Cd at Mn site is similar to that observed for ^{57}Fe at Mn site from Mössbauer measurements [8]. The $CeMn_2Ge_2$ compound shows a transition from paramagnetic to an antiferromagnetic ordering of the Mn atoms.

The Néel temperature T_N for the transition was estimated from our result to be ≈415 K while the transition temperature to the ferromagnetic ordering was estimated to be ≈320 K. These results are in better agreement with neutron dif-

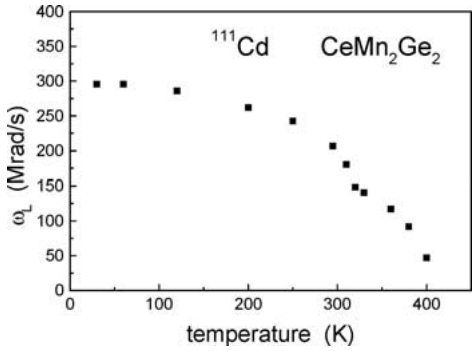

Figure 3. Temperature dependence of the Larmor frequency at the Mn site for $CeMn_2Ge_2$.

fraction measurements than the Mössbauer measurements most probably due to the influence of the high concentration of the Fe doping (∼5%) which may have caused small shifts in the transition temperature.

The result for the temperature dependence of the Larmor frequency of the ^{140}Ce at Ce site, resulting from the transferred magnetic hyperfine field from Mn moments, shows no magnetic interaction in the range of 320 K to 360 K. This is a consequence of the symmetry of the spin "up" and spin "down" arrangement of the Mn superlattice around the Ce site. Below 320 K we observed magnetic interaction in ^{140}Ce probe nuclei corresponding to the ferromagnetic ordering of the Mn moments. However, we have observed a sharp deviation from an expected Brillouin-like behavior below 100 K. Further measurements using ^{140}Ce probe as well as a careful analysis of this striking behavior will be the subject of a future publication.

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