

Magnetic interactions at Ce impurities in $\text{RE}\text{Mn}_2\text{Ge}_2$ (RE = La, Ce, Pr, Nd) compounds

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ABSTRACT

In the work reported in this paper, the temperature dependence of the magnetic hyperfine field (B_{hf}) at ^{140}Ce nuclei replacing Pr atoms in PrMn_2Ge_2 compound was measured by the perturbed angular correlation technique to complete the sequence of measurements in $\text{RE}\text{Mn}_2\text{Ge}_2$ (RE = La, Ce, Pr, Nd). Results show an anomalous behavior different from the expected Brillouin curve. A model was used to fit the data showing that the Ce impurity contribution (B_{hf}^{imp}) to B_{hf} is negative for NdMn_2Ge_2 below 210 K. The impurity contribution (B_{hf}^{imp}) at 0 K for all compounds is much smaller than that for the free Ce^{3+} , showing that the $4f$ band of Ce is more likely highly hybridized with $5d$ band of the host. Results show that direction of the localized magnetic moment at Mn atoms strongly affects the exchange interaction at Ce impurities.

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1. Introduction

Cerium ions play an important role in the diverse range of amazing magnetic properties of strongly correlated electron systems (SCES) having captured the attention of researchers focusing the investigation on magnetic interactions of Ce atoms with their electronic environment. Both, as an impurity or as a constituent element in rare-earth intermetallic compounds, Ce differs from the other rare-earth elements mainly by the location of the $4f$ state that is energetically very near the Fermi level. Consequently, the chemical and structural environment around Ce ions have a strong influence on the occupation number and strength of hybridization of $4f$ state with conduction electrons [1]. This feature enables a diversity of magnetic behaviors, such as different magnetic ordering, Kondo effect, strongly correlated electron effects, intermediate valence, etc., observed in compounds where Ce is an impurity or a native element.

Another interesting feature of Ce ions is that they can exist in intermediate valence (IV) states with values between +3 and +4 [2]. The IV states are, therefore, related to the occupation of the $4f$ band varying from a high population with a consequent localized spin moment found in the Ce ion, which creates a contribution from the Ce atom itself to the magnetic hyperfine field (B_{hf}) at Ce probe nuclei [3–5]. In the opposite extremity, the occupation of the $4f$ band has a low population due to de-localized $4f$ electrons

resulting in a very small contribution from the Ce atom electrons to the B_{hf} at Ce probe nuclei.

SCES have been studied using different experimental techniques such as neutron diffraction, magnetic susceptibility, and resistivity, etc. [6,7] Although these studies have resulted in a large volume of information, difficulties persist in understanding the behavior of $4f$ electrons of Ce in these systems. In particular, it is very difficult to experimentally observe magnetic interactions between Ce ions and magnetic atoms in systems where Ce is a dilute impurity in a magnetic host. In this respect, the study of hyperfine interaction at the Ce atom position in SCES systems can help to understand the physics behind their properties. In order to investigate the effect of different magnetic ordering on the exchange interaction between Ce ions and their magnetic environment it is important to have a series of compounds in which the magnetic ordering varies but the crystalline structure and the environment around the Ce impurity do not appreciably change. The series of compounds $\text{RE}\text{Mn}_2\text{Ge}_2$ (RE = rare-earth element) have attracted the attention due to different interesting physical properties such as heavy-fermion systems, magneto-caloric effect, superconductivity, etc. [8–11]. These compounds also present multiple magnetic transitions due to the presence of long-range ordering of manganese moments while the rare-earth sublattice (except for La and Ce) orders at low temperatures [12,13]. Recently, the magnetic hyperfine field at ^{140}Ce probe nuclei has been measured in LaMn_2Ge_2 [14], CeMn_2Ge_2 [15] and NdMn_2Ge_2 [16].

In the work reported in this paper, we have measured the temperature dependence of the magnetic hyperfine field (B_{hf}) at ^{140}Ce probe nuclei replacing Pr atoms in PrMn_2Ge_2 compound

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using the perturbed angular correlation (PAC) technique to complete the sequence of measurements in the compounds of the $\text{RE}\text{Mn}_2\text{Ge}_2$ series where RE is a light rare-earth. We, then, analyze the B_{hf} at ^{140}Ce nuclei in $\text{RE}\text{Mn}_2\text{Ge}_2$ (RE = La, Ce, Pr, Nd) in order to understand the different magnetic interactions presented by this series of compounds. We have fitted the experimental data with a model where Ce impurities are supposed to be in two states, one with low magnetic moment and another with high magnetic moment, in which the $4f$ electron of Ce is less or more localized, respectively.

2. Experimental methodology

PrMn_2Ge_2 samples were prepared by arc-melting the constituent elements (Pr = 99.9%, Mn, Ge = 99.999% purity) in the stoichiometric proportions in pure argon atmosphere. A 5 wt% excess of Mn was used because a small part of Mn was found to be lost during arc melting. After melting, the resulting ingot was sealed in an evacuated quartz tube and annealed at 800 °C during 24 h. The crystal structure of the compound was determined by X-ray diffraction (XRD) at room temperature. The results showed that the sample crystallized in the expected tetragonal structure of

ThC_2Si_2 in a single phase corresponding to the $I4/mmm$ space group. The resulting lattice parameters were $a = b = 4.126(3)$ Å and $c = 10.924(5)$ Å in agreement with previously reported values [17]. After structural characterization, a part of the sample was separated for the PAC measurements with ^{140}La (^{140}Ce) probe nuclei which were found to substitute Pr sites in the crystalline lattice. The other part of sample was used for magnetization measurements. The equipment used was a commercial Vibrating Sample Magnetometer (PPMS, Quantum Design) with an applied magnetic field of 100 Oe. Magnetization as a function of temperature was obtained using zero-field-cooled (ZFC) and field-cooled (FC) procedure.

For PAC measurements of B_{hf} at ^{140}Ce probe nuclei it is necessary to dope the compounds with an extremely dilute concentration of the radioactive parent ^{140}La nuclei. A small quantity of metallic La (replacing approximately 0.1 at% of Pr atoms), which was previously irradiated with neutrons in the IEA-R1 reactor of IPEN to produce radioactive ^{140}La , was added to part of the samples through arc melting followed by thermal annealing at 800 °C for 20 h under helium atmosphere. Measurements were carried out in the temperature range from 15 K to 330 K using a closed-cycle helium cryogenic device. Spin rotation spectra measured at some temperatures are displayed in Fig. 1.

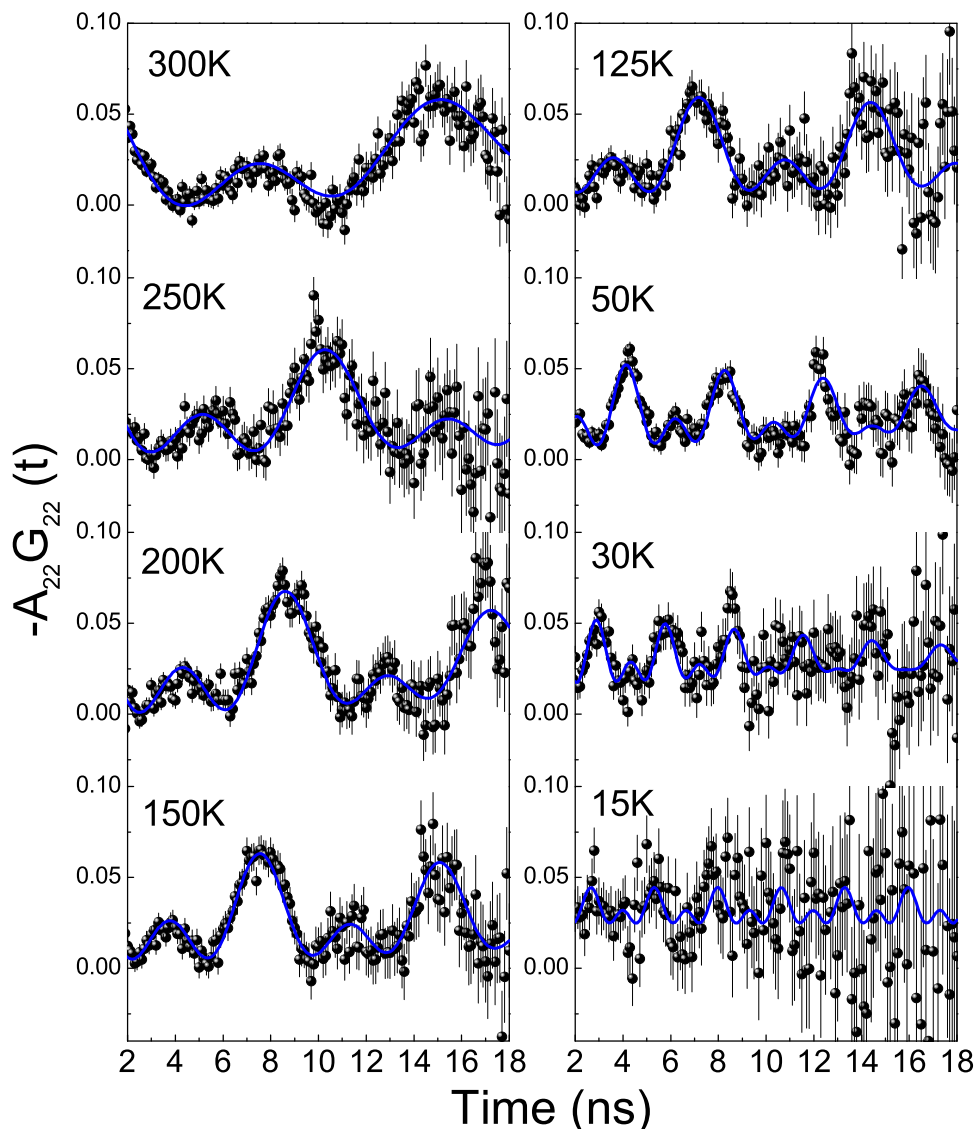


Fig. 1. Spin rotation spectra measured at indicated temperatures with ^{140}Ce at Pr sites in PrMn_2Ge_2 compound.

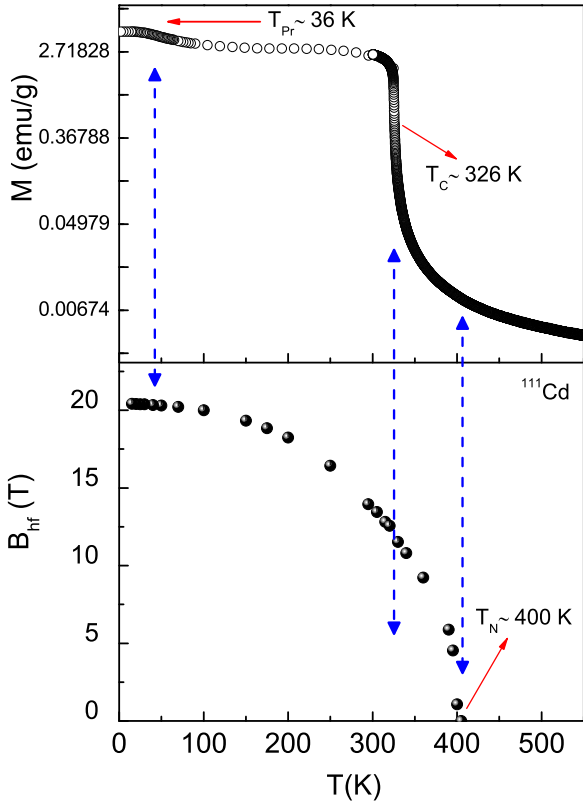


Fig. 2. Magnetization (top) and B_{hf} at ^{111}Cd probe substituting Mn positions (bottom) in a wide range of temperatures for PrMn_2Ge_2 compound [20].

The PAC method is based on the observation of hyperfine interaction of nuclear moments with extra nuclear magnetic field or electric field gradient (efg). A description of the experimental

method as well as details about the PAC measurements can be found elsewhere [15,18,19]. The perturbation factor $G_{22}(t)$ of the correlation function contains detailed information about the hyperfine interaction. Measurements of $G_{22}(t)$ allows, for magnetic dipole interaction, the determination of the Larmor frequency $\omega_L = \mu_N g B_{hf} / \hbar$, where μ_N is nuclear magneton, and straightforward calculation of the magnetic hyperfine field B_{hf} , provided that the g factor of the 487 keV energy level of ^{140}Ce is well known. The quadrupole moment Q of the 487 keV state as well as the time window (which depends on the half-life of the state) are relatively small and, therefore, the electric quadrupole interaction is too small to be observed.

3. Results and discussion

Magnetization measurements for PrMn_2Ge_2 with an applied field $H = 100$ Oe show several magnetic transitions with good resolution (see Fig. 2). The first transition from paramagnetic to anti-ferromagnetic phase was determined to occur at $T_N = 400$ K. The second transition from antiferromagnetic to ferromagnetic phase was determined at $T_C = 326.2$ K. The third transition corresponding to the Pr-4f spin sublattice ordering approximately at $T_{Pr} = 36$ K contributes to a slight increase in the magnetization. Measurements of NdMn_2Ge_2 magnetization also showed multiple magnetic transitions with good resolution [16]. The first transition occurs around 450 K and corresponds to the change from paramagnetic to anti-ferromagnetic phases. The second transition, from antiferromagnetic to ferromagnetic phases, is evident at 335 K. A third transition is observed to occur at T_t around 212 K and the fourth transition, associated to the magnetic coupling of Nd atoms, becomes apparent at $T_{Nd} = 25$ K. Previous results of B_{hf} measured with ^{111}Cd probe nuclei at Mn sites in REMn_2Ge_2 (RE = La, Ce, Pr, Nd) show a normal Brillouin-like temperature dependence with small discontinuities in the curve at transition temperatures [15,16,20,21].

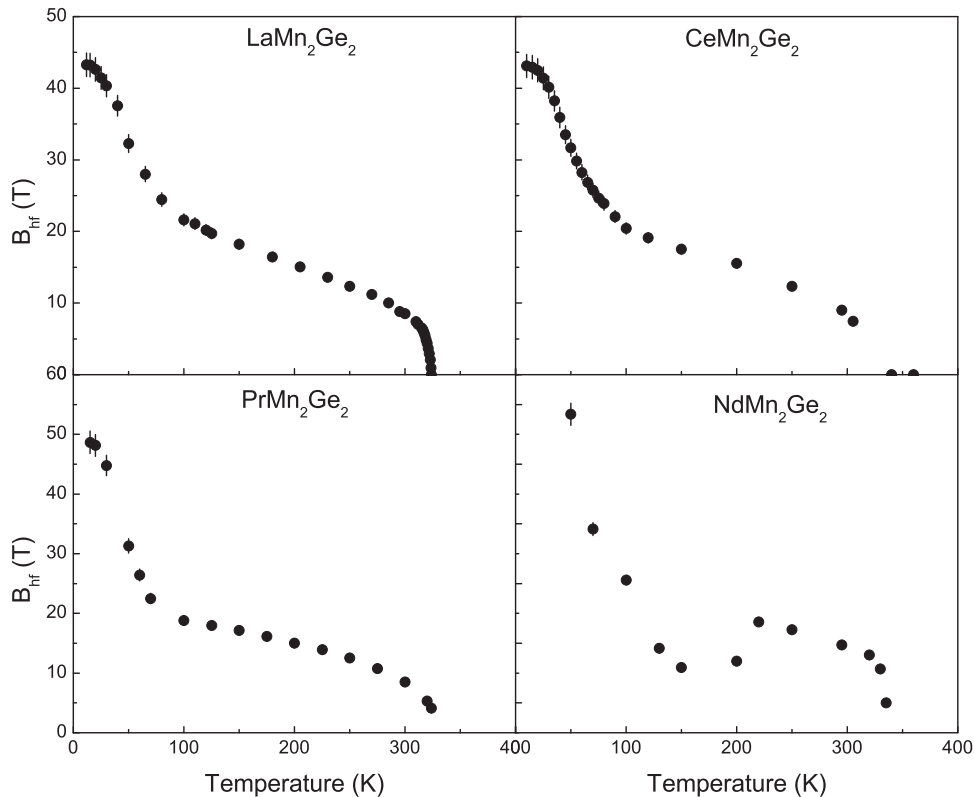


Fig. 3. Temperature dependence of B_{hf} measured at ^{140}Ce at RE sites in REMn_2Ge_2 compound.

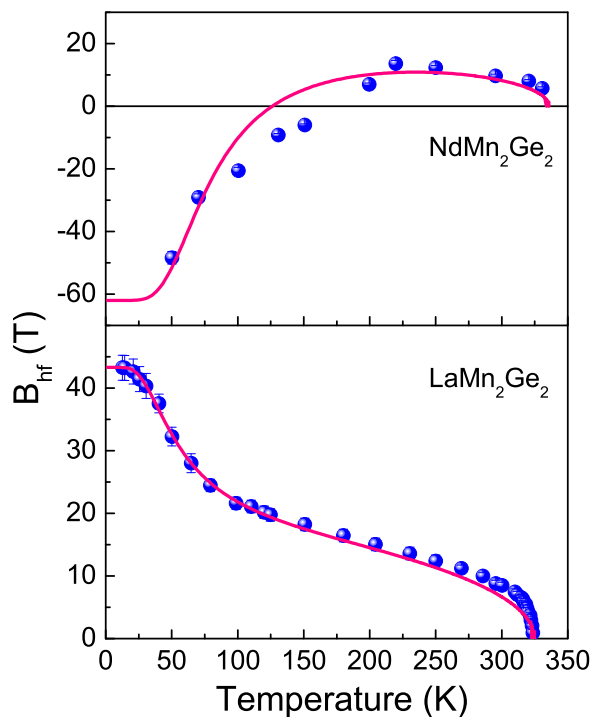


Fig. 4. Fit of the model (represented by the solid red curves) described in the text to experimental data of the temperature dependence of the magnetic hyperfine field measured at ^{140}Ce in NdMn_2Ge_2 (top) and LaMn_2Ge_2 (bottom). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The temperature dependence of B_{hf} at ^{140}Ce probes in REMn_2Ge_2 compounds, displayed in Fig. 3, shows, however, an anomalous behavior in all cases. This behavior has been attributed to the contribution of the $4f$ band of the Ce impurity, which is located very near the Fermi level [15]. For NdMn_2Ge_2 one can observe a more complex behavior in which a decrease of the B_{hf} around 210 K is clearly found because of the reorientation of the Mn spins direction from the c -axis to the ab -plane, which is also evidenced in the magnetization measurements.

In order to analyze the different contributions to B_{hf} measured with ^{140}Ce , we adapted a model developed by Campbell [22] in which the Ce magnetic impurity is considered to be in two magnetic moment states: a high moment state with well-localized band and a low moment state with an almost delocalized band. In this two-state model, a magnetic hyperfine field at impurity nuclei (^{140}Ce) is associated to each state. These fields correspond to the host contribution (low moment state) and to the Ce ion contribution (high moment state) to the magnetic hyperfine field at ^{140}Ce . These fields are those considered in another model used to fit anomalous temperature dependence of B_{hf} at ^{140}Ce observed in compounds where the magnetic ions are rare-earth ions and Ce impurities are located at the rare-earth sites [3]. In the present work, the magnetic ion is a transition metal ion and the model used by Cavalcante et al. [3], where the value of each field contributing to the effective magnetic hyperfine field are directly used in the mathematical expression, did not fit well the experimental data. Contrarily to rare-earth ions, in which $4f$ -bands are more localized and therefore do not interfere in the energy of $4f$ -electron of Ce, transition elements are much less localized and their $3d$ -bands can hybridize with the $4f$ -band of Ce. Consequently, the energy approach seems to be the better way. The observed anomalous behavior of the temperature dependence of B_{hf} is associated with a transition from the well localized to the

delocalized moment state whose energy difference depends on the temperature with the difference in energy between the two states at 0 K given by E_0 . The curve fitted to experimental data is shown in Fig. 4 for NdMn_2Ge_2 and LaMn_2Ge_2 . Although the sign of the magnetic hyperfine field was not measured, we consider that the sign of B_{hf} for NdMn_2Ge_2 change direction for values below around 210 K and it is opposite to the sign of B_{hf} in the same range shown in Fig. 3. This assumption is based on the model described by Torumba et al. [23] in which the $3d$ moments of the transition metal couple antiferromagnetically with the $5d$ and $4f$ moments of the rare-earth atoms. The temperature behavior of NdMn_2Ge_2 is then displayed in Fig. 4 with negative values for B_{hf} below 200 K.

From the parameters obtained from the fit it is possible to calculate two contributions to B_{hf} measured at ^{140}Ce , that from the magnetic host ($B_{\text{hf}}^{\text{host}}$) and another from the probe itself ($B_{\text{hf}}^{\text{imp}}$). Results from the fit are displayed in Table 1.

From the fit of the model to the experimental data of the temperature dependence of B_{hf} measured with ^{140}Ce , one can observe that B_{hf} at $T = 0$ K increases from La to Nd, which is due to the decrease in the RE-Mn distance from 3.453 Å for LaMn_2Ge_2 to 3.410 Å for NdMn_2Ge_2 . B_{hf} for NdMn_2Ge_2 has opposite sign and is much higher than values for the other compounds, which is ascribed to the coupling between Nd atoms below 25 K. Nd compound also presents a change in the direction of the Mn spins in the ferromagnetic ordering. Below T_c it is along the c -axis and below T_i a reorientation occurs leaving the direction of Mn spins along the ab -plane. Results show that this reorientation changes the sign of the impurity contribution $B_{\text{hf}}^{\text{imp}}$ to the opposite direction, which is clearly seen by the sudden decrease observed in the $B_{\text{hf}} \times T$ curve around 210 K. When the temperature decreases, the absolute value of B_{hf} continues to increase when T decreases reaching the highest absolute value observed due to the coupling of Nd atoms. At this point, it is noteworthy that PAC results using ^{140}Ce probe nuclei can follow all this complex magnetic behavior shown by this compound. The temperature dependence of B_{hf} for PrMn_2Ge_2 shows a behavior slightly different from those for LaMn_2Ge_2 and CeMn_2Ge_2 with a steeper increase of the B_{hf} values at low temperatures, which was ascribed to the coupling of Pr atoms. This increase is not as sharp as that for NdMn_2Ge_2 probably due to the larger distance between RE atoms. All values of $B_{\text{hf}}^{\text{imp}}$ at $T = 0$ K are much smaller than that for the free Ce^{3+} ion, which is around 183 T [24], showing that the $4f$ band of Ce impurity is more likely highly hybridized with $5d$ band of the host. Results suggest that the magnetic exchange interaction of Ce impurity with the magnetic ions is strongly dependent of the direction of the localized magnetic moments in these ions from the host. This dependence was also observed for B_{hf} at ^{111}Cd at In sites in antiferromagnetic CeIn_3 compound in which B_{hf} should be vanish at In sites [25,26].

Table 1

Parameters obtained from the fit of the two-state model (see text). Values of B_{hf} in Tesla (T) are given at 0 K.

Compound	T_c (K)	E_0 (meV)	B_{hf}	$B_{\text{hf}}^{\text{host}}$	$B_{\text{hf}}^{\text{imp}}$
LaMn_2Ge_2	323	10.6	43.3	9.0	34.3
CeMn_2Ge_2	320	9.1	43.8	10.1	33.7
PrMn_2Ge_2	336	8.7	49.5	10.9	38.6
NdMn_2Ge_2	320	8.5	-62.0	32.0	-94.0

4. Conclusions

In the work reported in this paper we have investigated the temperature dependence of the magnetic hyperfine field at ^{140}Ce impurities in REMn_2Ge_2 ($\text{RE} = \text{La, Ce, Pr, Nd}$) compounds. We have supposed that Ce ions can be at two states as impurity in these compounds: a high moment state, where $4f$ electrons are more localized and a low moment state with $4f$ electrons not well localized. As B_{hf} is associated to each state, the competition between these two states will shape the temperature dependence of B_{hf} . The magnetic hyperfine field at ^{140}Ce reflects the magnetic exchange interaction between Ce ion and its magnetic environment and the results show that the magnetic exchange interaction of Ce impurity with the magnetic ions is strongly dependent of the direction of the localized magnetic moments in these ions of the host.

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