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Study of hyperfine interactions in GdIn₃

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In the present work, an experimental and theoretical study of the hyperfine interactions at Gd and In sites in GdIn₃ were performed. The experimental measurements were carried out by perturbed angular correlation spectroscopy using ¹⁴⁰Ce and ¹¹¹Cd nuclear probes substituting Gd and In sites, respectively. Results for ¹¹¹Cd probe at In sites in GdIn₃ revealed only electric quadrupole interactions, differently from the results for CeIn₃ where, in addition to quadrupole interactions, a magnetic hyperfine field (B_{hf}) was also observed at In sites. The temperature dependence of B_{hf} at ¹⁴⁰Ce on Gd sites in GdIn₃ could be fitted by a Brillouin curve, and the extrapolated B_{hf} value to 0 K was found to be much smaller than that at ¹⁴⁰Ce in CeIn₃. *Ab-initio* electronic structure calculations for GdIn₃ matrix doped with Ce were confronted with experimental data in order to explain such differences. The calculations were carried out within density functional theory using Augmented Plane Waves plus local orbitals basis functions as embodied in the WIEN2K package and with the GGA + U approximation. The value for the Hubbard U parameter was determined for each case. Results of the calculations show that the absence of B_{hf} in Cd probes in GdIn₃ is related with the orientation of the magnetic moments in (001) direction, whereas in CeIn₃ the magnetic moments are oriented out of this direction. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4797624>]

I. INTRODUCTION

REIn₃ intermetallic compounds are systems in which the rare-earth (RE) elements are immersed in a sea of *sp*-type conduction electrons, which make them very good systems for the study of magnetism in rare-earth compounds. In recent decades, such systems have been studied by means of different experimental methods as well as theoretical calculations in order to determine their magnetic properties.^{1,2} In particular, REIn₃ has been the object of *ab-initio* calculations aiming to describe the nature of the electric field gradient (EFG) and its dependence on the 4*f* electrons located at the rare-earth ions. The magnetic hyperfine field (B_{hf}) acting at the rare-earth and at the In positions, on the other hand, has been much less explored in such studies.² Among the REIn₃ compounds, it is worth to highlight the compound CeIn₃, which has been extensively studied because it shows characteristics that are important to the understanding of strongly correlated systems. It shows Kondo-type as well as superconducting behavior when subjected to an externally applied pressure.^{3–5} These phenomena are attributed to the nature of the interaction between the single 4*f*-electron in Ce atom with conduction electrons. However, little has been discussed regarding the origin and behavior of B_{hf} in these compounds and there are some differences between experimental results and theoretical calculations.^{2,6}

In this paper, we report an investigation of the behavior of the hyperfine parameters as a function of temperature in the GdIn₃ by means of perturbed angular correlation (PAC) spectroscopy using highly diluted ¹⁴⁰Ce and ¹¹¹Cd probe nuclei located at gadolinium and indium atomic positions, respectively. The results are compared with those of CeIn₃

(Ref. 8) and with the first-principles electronic structure calculations in order to have a better comprehension of the mechanisms involved in the magnetic properties of these compounds.

II. EXPERIMENTAL DETAILS

The samples of GdIn₃ were prepared by arc melting the constituent metals Gd (99.9%) and In (99.9999%) in stoichiometric proportions. The PAC probes were introduced by remelting the samples in the arc furnace with the addition of: (1) small quantity (<0.1%) of natural La irradiated with neutrons in the IEA-R1 reactor at IPEN for 12 h at the neutron flux of $3 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ that produces the ¹⁴⁰La isotope; (2) a few drops of carrier free ¹¹¹In isotope in the form of InCl₃ solution. The sample, before addition of the radioactive probes, was analyzed by X-ray diffraction, which confirmed a single phase corresponding to the AuCu₃-type structure. PAC measurements were carried out with a four BaF₂ detector spectrometer using ¹⁴⁰La(¹⁴⁰Ce) and ¹¹¹In(¹¹¹Cd) nuclear probes. The gamma cascades of 171–245 keV in ¹¹¹Cd populated in the electron capture decay of ¹¹¹In and 329–487 keV in ¹⁴⁰Ce populated from the β^- decay of ¹⁴⁰La were used for the measurements. The experimental details and methodology used can be found elsewhere.⁷ The measurements were carried out in the temperature range of 12–300 K using a closed cycle helium cryogenic system.

III. RESULTS AND DISCUSSION

Some of the PAC spectra measured at different temperatures and their respective Fourier transforms are shown in Fig. 1 for ¹¹¹Cd probe at In sites in GdIn₃ compound. The results of measurements in the temperature range of 15–300 K showed a single well-defined quadrupole frequency (ν_Q) with

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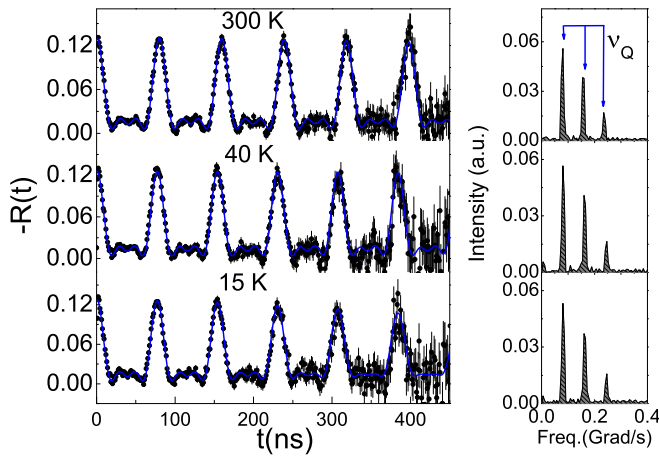


FIG. 1. PAC perturbation functions and respective Fourier transforms for $^{111}\text{In}(^{111}\text{Cd})$ probe at different temperatures in GdIn_3 . Solid lines are the least squares fit to the theoretical function.

an asymmetry parameter $\eta = 0$. No magnetic interaction was observed even below the known anti-ferromagnetic transition temperature, $T_N \sim 45$ K.

PAC spectra using ^{140}Ce probe nuclei, which replace Gd positions in GdIn_3 , are shown in Fig. 2. These spectra were fitted using a model in which only magnetic dipole interaction is considered once the quadrupole moment (Q) of the ^{140}Ce intermediate state involved in the gamma cascade used, is known to be very small. As a consequence, only magnetic interactions are observed with ^{140}Ce below $T_N = 45$ K for GdIn_3 .

In Figure 3(a), one can observe the behavior of quadrupole frequency ν_Q , measured with ^{111}Cd in GdIn_3 , as a function of temperature (T) compared with a similar result for CeIn_3 .⁸ The experimental values of ν_Q could be fitted by the equation: $\nu_Q(T) = \nu_Q(0)(1 - \beta T^{3/2})$, which shows that ν_Q follows a $T^{3/2}$ dependence. The quadrupole frequency at 0 K obtained from the fit for CeIn_3 is $\nu_Q(0) = 81.8(1)$ MHz, which corresponds to a major component of the electric field gradient at Cd impurities, $V_{zz}^{Cd} = 4.08 \times 10^{21} \text{V/m}^2$. In the

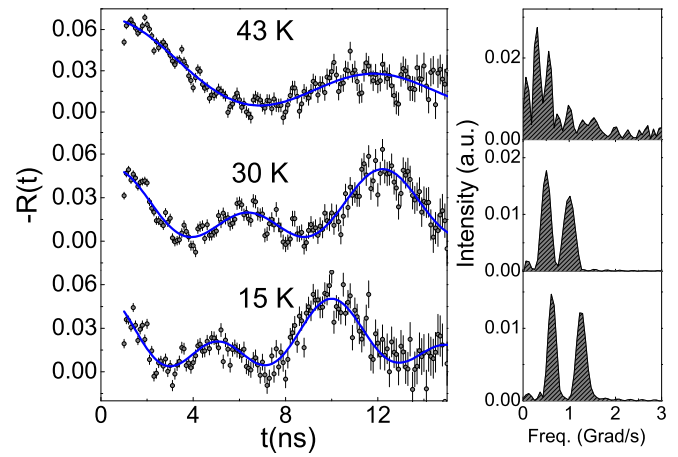


FIG. 2. PAC perturbation functions for ^{140}Ce in GdIn_3 at indicated temperatures and their respective Fourier transforms. The solid lines are the least squares fit to the theoretical function.

case of GdIn_3 , $\nu_Q(0) = 87.1(1)$ MHz and corresponds to $V_{zz}^{Cd} = 4.34 \times 10^{21} \text{V/m}^2$. This value is in very good agreement with the value of $4.3 \times 10^{21} \text{V/m}^2$ obtained from the 4f open-core Density Functional Theory (DFT) calculations by Asadabadi *et al.*²

The results of PAC measurements with ^{111}Cd probe in GdIn_3 below T_N did not show any magnetic interaction, in contrast to the results for CeIn_3 measured with the same probe which show the existence of a magnetic interaction varying with temperature below T_N .⁸

For ^{140}Ce probes in CeIn_3 and GdIn_3 , the temperature dependence of B_{hf} follows a Brillouin-type function with total angular momentum $J_{Ce} = 5/2$ and $J_{Gd} = 7/2$, respectively, with magnetic transition temperatures $T_N = 10$ K and $T_N = 45$ K (Fig. 3(b)).

Electronic structure calculations were carried out using DFT with “Augmented Plane Waves plus local orbitals” (APW + lo) basis functions as implemented in the WIEN2K package.⁹ The “Local density approximation” (LDA) and “Generalized Gradient Approximation” (GGA) were

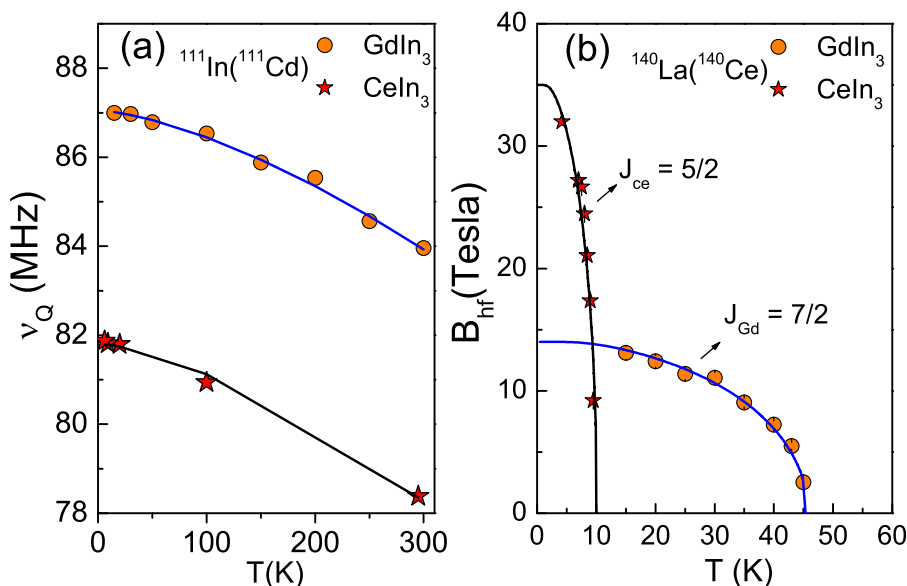


FIG. 3. Temperature dependence of the quadrupole frequency ν_Q at In sites (a) and temperature dependence of the magnetic hyperfine field at Ce probes (b) for both GdIn_3 and CeIn_3 . Data for CeIn_3 were taken from Ref. 8. The solid lines in (b) represent the Brillouin function for total angular momentum $J_{Ce} = \frac{5}{2}$ and $J_{Gd} = \frac{7}{2}$.

TABLE I. Magnetic hyperfine field and magnetic moments on Gd in GdIn₃ (Gd rows) and on Ce atoms substituting for Gd in GdIn₃ (Ce rows), as determined from first-principles electronic structure calculations. B_{hf} (orbital, spin-dipolar and Fermi-contact) in Tesla and magnetic moments (spin and total components) in bohr-magnetons.

	B_{orb}	B_{dip}	B_c	B_{tot}	μ_S	μ_{tot}
Gd:GdIn ₃ -GGA	9.48	-0.15	37.0	46.3	6.90	6.85
Gd:GdIn ₃ -GGA + U	1.12	-0.43	39.4	40.1	7.06	6.87
Gd:GdIn ₃ -LDA	13.4	-0.04	-0.52	12.8	6.85	6.84
Ce:GdIn ₃ -GGA	-22.4	-0.32	0.55	-22.1	0.67	0.25
Ce:GdIn ₃ -GGA + U	-30.7	-1.02	2.86	-28.9	0.91	0.33
Ce:GdIn ₃ -LDA	-25.0	-0.46	-2.22	-27.7	0.58	0.12

employed for the exchange and correlation functional.¹⁰ Volume optimizations were done with spin-polarized calculations simulating the ($\pi\pi\pi$) anti-ferromagnetic structures. The Brillouin-zone integrations were performed with 2000 k-points distributed over a tetrahedral mesh. The number of basis functions was defined by $R_{min}K_{max} = 8$, which determines the plane-wave energy cutoff of 140 eV (see details in Ref. 9). The Ce impurity in GdIn₃ was simulated by a $2 \times 2 \times 2$ super-cell where one Gd atom is substituted by Ce. Spin-orbit effects were introduced as second variational step with scalar-relativistic wave-functions and with the orientation of magnetization into the (001) and (111) directions. To improve the representation of 4f states of the rare-earths, a Hubbard term with an effective energy U_{eff} is added to the Hamiltonian. The U_{eff} was determined under the prescriptions of Madsen and Novák¹¹ and resulted 4.90 eV and 6.53 eV for Ce and Gd, respectively.

From these calculations, it is observed that the B_{hf} on In sites is sensitive to the direction of magnetization. For CeIn₃, the magnetization direction (111) allows for a net B_{hf} on In sites due to a small polarization of In p orbitals (see Ref. 5). The same result is obtained for GdIn₃. However, since this result is contrary to the experimental evidence in the present work, we confirm that the magnetization direction in GdIn₃ is (001) since the B_{hf} is exactly canceled by symmetry in this case. These calculations further show that the electric field gradient at In site is not sensitive to the changes of the magnetic direction from (001) to (111) in both the compounds, CeIn₃ and GdIn₃.

Part of the results obtained from the calculations are presented in Table I, which shows values for B_{hf} on Gd sites in GdIn₃ (Gd:GdIn₃) and values for B_{hf} on Ce probes at Gd sites in GdIn₃ (Ce:GdIn₃). With the GGA functional, the B_{hf} for Gd is predominantly due to the Fermi-contact interaction, whereas for Ce in GdIn₃, the main contribution comes from the orbital field. On the other hand, the LDA approximation results with a larger orbital contribution and a negative Fermi-contact interaction in both the Gd and Ce probes.

The experimental value for B_{hf} at Gd is 23.7(5) T (see Ref. 12), whereas the correspondent value at Ce is 34.9 T (Figure 3(b)). Thus, the LDA functional underestimates, whereas the GGA (and GGA + U) overestimates the B_{hf} at Gd in GdIn₃. Since Gd ion has a pure S state, no orbital contribution is expected. This situation is correctly reproduced by the GGA + U functional. Indeed, at the Ce probes, both the LDA and GGA + U functionals result in similar agreement with total experimental B_{hf} .

Comparing the magnitude of B_{hf} at Ce in CeIn₃ (see Ref. 5) with that at Ce in GdIn₃, one observes that the theoretical results follow the experimental trend of decreasing the B_{hf} at Ce on going from CeIn₃ to GdIn₃ (see Figure 3(b)), but only to a small extent, going from 25.05 T (see Ref. 5) to 22.1 T, as compared to the experimental larger reduction from 34.9 T to 13.9 T. Moreover, one can see that the B_{hf} components at Ce are very similar in CeIn₃ and GdIn₃. Thus, from the theoretical DFT results, it can be concluded that the Ce probes behave very similarly, independently whether the host is CeIn₃ or GdIn₃. Such behavior could be understood if the 4f ground state of Ce would be determined exclusively by the charge distribution around the Ce ion, as predicted in the point charge model: the charge distributions are expected to be very similar in CeIn₃ and GdIn₃. On the other hand, the spin-polarizations of the conduction electrons in both the compounds are very different (see the values of the magnetic moments in Table I). This is certainly the reason for the different values of B_{hf} at Ce in CeIn₃ and GdIn₃.

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