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Magnetic hyperfine field at highly diluted Ce impurities in the antiferromagnetic compound $GdRh_2Si_2$ studied by perturbed gamma–gamma angular correlation spectroscopy

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ABSTRACT

The magnetic properties of $GdRh_2Si_2$ compound were investigated by hyperfine interactions and magnetization measurements. The temperature dependence of the magnetic hyperfine field (mhf) at highly diluted ^{140}La–^{140}Ce probe nuclei in the $GdRh_2Si_2$ compound was measured using perturbed gamma–gamma angular correlation spectroscopy. A well-defined magnetic interaction is observed at ^{140}Ce substituting Gd atoms in the compound below the Néel temperature (T_N). However, the temperature dependence of mhf shows a deviation from the expected Brillouin-like behavior and a sharp increase of mhf values is observed for temperatures below approximately half of T_N. This behavior has been associated with an additional magnetic interaction which is ascribed to the polarization of Ce spin moment induced by the magnetic field coming from Gd ions. A model based on the molecular-field theory is used to fit the temperature dependence of the mhf. The mhf contributions from the probe atom itself and from the magnetic Gd ions of the host matrix are determined. The results are compared to previous measurements of mhf at ^{140}Ce in Gd and GdAg compound. The contribution from the host to the mhf is discussed in terms of the f–s and f–d interactions.

1. Introduction

In order to study the origin and understand a wide variety of interesting magnetic phenomena associated with rare-earth intermetallic compounds it is useful to investigate the magnetic field from an atomic scale in an attempt to identify the origin of such phenomena. For instance, in a fundamental aspect, the way 4f spins of rare-earth elements couple each other in many intermetallic compounds is not well determined yet. Among the most powerful tools used to study electric and magnetic fields on an atomic scale are the hyperfine interactions (hfi), which are the interactions between the nuclear moments and magnetic fields or electric field gradients due to the electronic charge outside the nucleus. Experimental measurement of magnetic hyperfine interactions at a rare-earth site requires probe nuclei occupying the rare-earth positions. These probe nuclei can be the rare-earth ions themselves present in the studied compounds when resonance techniques such as Mössbauer spectroscopy or nuclear magnetic resonance (NMR) are used, or some different nuclei which can probe hyperfine interactions. Perturbed gamma–gamma angular correlation (PAC) spectroscopy is a hyperfine interaction technique which uses radioactive probe nuclei introduced in the samples substituting specific atoms in the crystal lattice. PAC spectroscopy therefore allows the use of the same probe nucleus to investigate magnetic hyperfine interactions in a family of compounds with different rare-earth elements.

A quite convenient probe nucleus for PAC measurements in rare-earth compounds is ^{140}Ce which can be used to investigate the local magnetism in the compounds where it substitutes rare-earth ions. Among the rare-earth elements, cerium atoms are responsible for interesting physical phenomena, such as superconductivity, Kondo effect, striking magnetic behavior and intermediate valence state, which are solely due to its single 4f electron with energy very close to the Fermi level that makes the Ce ion very sensitive to the chemical environment. Ce ions can exist in two valence states, one of them is Ce^{3+} where only one 4f electron remains and consequently a localized spin moment is found in such ions. These ions also create a contribution from the probe itself to the magnetic hyperfine field (mhf). The other valence state is Ce^{4+} with the electronic configuration of xenon and therefore no 4f electron is present, and as a consequence no contribution from the probe electrons to the mhf exists. Ce is also known to present intermediate valence configuration in many compounds [1,2]. The 4f spin in the Ce^{3+} configuration or in the intermediate valence configuration of Ce ions of ^{140}Ce probe nuclei can, therefore contribute to the mhf and interfere with

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the measurements of the mhf from the magnetic host atoms. The magnetic hyperfine field at the probe nuclei of magnetic rare-earth ions diluted in magnetic intermetallic compounds is however, in itself a very interesting and yet not well understood subject mainly because the exchange interaction between the localized magnetic moment on the probe atom and the magnetic host ions is not well described. It is noteworthy however, that the localized moment on diluted magnetic ions in magnetic matrices can only be measured by local techniques such as hfi techniques.

The problem of localized moments formation at impurities in metals was intensively studied in the past, particularly for magnetic transition metal impurities in non-magnetic metals [3], chiefly after the discovery of the Kondo effect [4], which describes the scattering of conduction electrons in a metal due to magnetic impurities in order to explain the temperature dependence of electrical resistivity. However, when the metal host is magnetic, an interaction between the host ions and the magnetic impurities appears and the situation is more complex and not unambiguously described yet. There are several measurements of mhf at 3d impurity probes in magnetic transition elements or compounds (see, for instance, references [5–7]), however very few studies on mhf measurements at rare-earth probes in rare-earth elements or compounds have been reported [8–10].

In the present work we have measured the temperature dependence of the mhf at ^{140}Ce in the intermetallic compound $GdRh_2Si_2$. Using a model based on molecular field theory [11] with some modifications [12], we have calculated the two contributions to the mhf: from the probe ions themselves and that from the magnetic Gd ions of the host matrix. The results are compared to those obtained from the fit to the temperature dependence of mhf at diluted ^{140}Ce probes in pure Gd with $T_C = 293$ K [8] and GdAg with $T_N = 132$ K [13] reported previously. These results along with the results from magnetization measurements were used to investigate the coupling between the 4f spins of Gd ions in terms of f–s and f–d interactions.

$GdRh_2Si_2$ belongs to the family of compounds RRh_2Si_2, where R is a rare-earth element, which have been extensively studied in the past since the discovery of superconductivity in $CeCu_2Si_2$ by Steglich et al. [14]. RRh_2Si_2 compounds have attracted special interest due to its several physical properties such as, for instance, long range magnetic ordering, antiferromagnetism, superconductivity, and Kondo effect [15]. Differently from the rest of the family, $GdRh_2Si_2$ has a relatively high Néel temperature ($T_N = 106$ K) ascribed to the strong exchange interaction between Gd ions whose localized magnetic moments ($\mu = 8.22\mu_B$)are oriented in the *a*–*b* plane, as revealed by Mössbauer measurements [16].

$GdRh_2Si_2$ crystallizes in the $ThCr_2Si_2$ prototype structure which belongs to the spatial group I4/*mmm* [17]. In this structure rare-earth atoms form a body-centered tetragonal sub-lattice. In such structure, rare-earth atoms occupy the crystallographic position (2a):(0,0,0), Si atoms occupy positions (4e):(0,0,z);(0,0,$\bar{z}$) while Rh atoms occupy the position (4d):(0,1/2,1/4).

2. Experimental

The samples of $GdRh_2Si_2$ were prepared by repeatedly melting the stoichiometric quantities of pure elements (Gd 99.9%, Rh 99.99% and Si 99.9999%) under argon atmosphere purified with a hot titanium getterer in an arc furnace. Subsequently, the samples were sealed under vacuum and annealed for one week at 900 °C. Afterwards, the samples were cut into slices and a part was used for the structural characterization with X-ray diffraction (XRD) technique using Cu K_α X-rays. The resulting XRD data were analyzed using the Rietveld refinement method. After assuring the correct crystalline phase, the remaining part of sample was again arc-melted but this time with radioactive ^{140}La nuclei, obtained by neutron irradiation of lanthanum metal, to substitute about 0.1% of Gd atoms. Radioactive ^{140}La ($t_{1/2} = 40.2$ h) β^- decays to the excited states of ^{140}Ce and a known gamma cascade in this decay is used for the PAC measurement.

An interesting feature of ^{140}Ce probe nuclei is that the intermediate state of the gamma cascade used in the PAC measurements has a relatively small electric quadrupole moment ($Q = 0.35$ b [18]), and a short half-life of only 3.5 ns which, limits the time window for the observation of hyperfine interactions. This implies that only quadrupole interactions with frequencies much higher than those usually found in most materials would be observed as a consequence, only magnetic dipole interactions might be observed with ^{140}Ce probe nuclei.

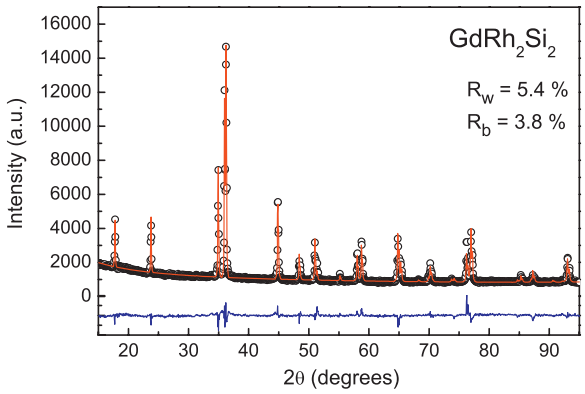

Fig. 1. X-ray diffraction pattern of $GdRh_2Si_2$ sample doped with approximately 0.1% of La. The solid line represents the calculated pattern obtained from the Rietveld refinement method. The residuals are shown in the lower part of each curve.

Samples used for PAC measurements with diluted radioactive ^{140}La were annealed under vacuum for 72 h at 950 °C, and subsequently measured in a conventional fast–slow coincidence set-up with four conical BaF_2 detectors. The PAC measurements were carried out in the temperature range of 10–295 K by using a closed-cycle helium cryogenic device. 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 at Gd sites. The time resolution of the system was about 0.6 ns for the ^{140}Ce gamma cascade.

The PAC method is based on the observation of hyperfine interaction of nuclear moments with extra-nuclear magnetic field or electric field gradient. A detailed description of the method can be found elsewhere[19,20]. For an unpolarized ferromagnetic sample consisting of randomly oriented domains, the perturbation factor $G_{22}(t)$ of the correlation function, which contains detailed information about the hyperfine interaction, can be written (neglecting the A_{44} terms) as:

$$R(t) = A_{22}G_{22}(t) = A_{22}[0.2 + 0.4\cos(\omega_L t) + 0.4\cos 2(\omega_L t)] \quad (1)$$

Measurement of $G_{22}(t)$ allows the determination of Larmor frequency $\omega_L = \mu_N g B_{hf}/\hbar$ and since the *g*-factor of the intermediate level is well known it is possible to determine the magnetic hyperfine field B_{hf}. The experimental data at temperatures below T_N were analyzed with a pure magnetic dipole interaction.

The magnetization measurements of the ^{140}La-doped sample were performed after the radioactive isotope had mostly decayed. Those measurements were carried out with a commercial vibrating sample magnetometer (VSM), in the temperature range of 4.2–300 K with an externally applied magnetic field of 0.5 T.

3. Results and discussion

The results of XRD measurements indicated the formation of a single phase with tetragonal crystalline structure of $ThCr_2Si_2$-type which belongs to the I4/*mmm* space group. Within the resolution of XRD, no secondary phases were observed as can be seen in Fig. 1. The lattice parameters obtained from the analysis were $a = 4.041$ Åand $c = 9.981$ Å.

In Fig. 2 is shown the DC magnetic susceptibility as a function of the temperature. The features observed in the figure suggest that the sample shows an antiferromagnetic order with a Néel temperature of $T_N = (106 \pm 1)$ K. Below T_N the susceptibility shows an almost constant behavior with a slight tendency to increase as the temperature decreases to 4.2 K. Above the transition temperature, the susceptibility shows the characteristic Curie–Weiss behavior of the paramagnetic state as the temperature is increased. The fitting of the Curie–Weiss law to the experimental data yielded an effective magnetic moment $\mu_{eff} = 8.44(2)\mu_B$ per Gd ion. This value is somewhat higher than the calculated value considering the spin $S = 7/2$ ($7.94\mu_B$). This difference according to Czjzek et al. [16] is due to an additional magnetic moment induced at the 5d-electrons level by the 4f–5d exchange interaction which is added to the magnetic

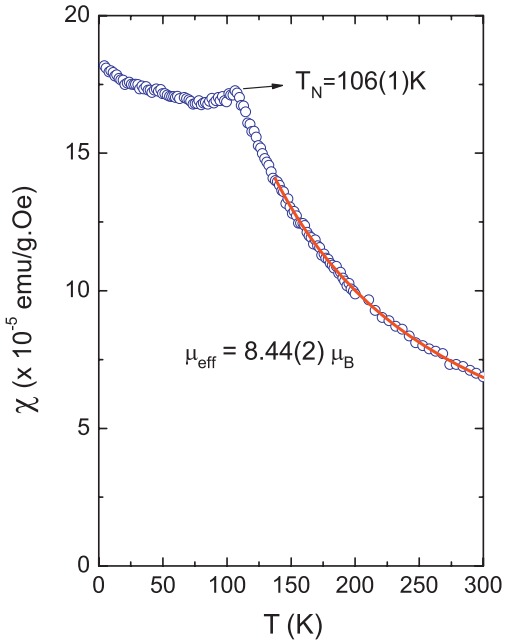

Fig. 2. Magnetic susceptibility ($\chi(T)$) for $GdRh_2Si_2$ compound obtained with an external magnetic field of 0.5 T. The transition temperature at 106 K is clearly observed. The solid line represents the fit of experimental data to the Curie–Weiss function.

moment of 4f-electrons of Gd ions. As a consequence, the effective magnetic moment is expressed by: $\mu_{eff} = \mu^{4f} + \mu^{5d}$ [16].

Fig. 3 shows some of the PAC spectra for $GdRh_2Si_2$ measured at temperatures below T_N using the $^{140}La \rightarrow {}^{140}Ce$ probe. Solid curves represent the least squares fit of the expression 1 to the experimental data. Results of the fit indicated a unique Larmor frequency ω_L with a small distribution ($\delta \sim 2\%$). In order to calculate the magnetic hyperfine field B_{hf} a g-factor value of $g = 1.014(38)$ was used for the 486 keV intermediate level of the gamma cascade in ^{140}Ce [21].

Fig. 4a shows the temperature dependence of the hyperfine field B_{hf} at ^{140}Ce in $GdRh_2Si_2$. The observed behavior corresponds to the antiferromagnetic ordering of the Gd moments. Below the Néel temperature, the B_{hf} values are expected to show a normal Brillouin type behavior. However, the observed data at temperatures below about 40 K show an anomalous behavior with a sharp deviation from the expected curve described by the standard Brillouin function for an antiferromagnet.

The temperature dependence of the hyperfine field near the transition temperature was also investigated. Some of the B_{hf} values immediately below T_N were fitted to the modified Curie–Weiss law: $B(T) = B(0)(1 - T/T_N)^{\beta}$, where β is an exponent. The resulting value of T_N obtained from this fitting was 106 ± 1 K, which completely agrees with that obtained from magnetization measurements. The fit also yielded a value of 0.38(4) for the exponent β, which is close to the theoretical value of the critical exponent expected for a 3-dimensional isotropic Heisenberg magnet ($\beta_H = 0.365(1)$) [22].

The observed unusual temperature dependence of B_{hf} measured at ^{140}Ce in $GdRh_2Si_2$ is quite similar to that already seen in Gd [8] and GdAg [13]. In Fig. 4b the experimental data taken from the literature is shown, where B_{hf} is plotted as a function of reduced temperature T/T_0, T_0 being the magnetic ordering temperature. Interestingly, it is observed that the behavior of B_{hf} as a function of temperature is quite similar for the three compounds. Near the ordering temperature T_0, just below it, B_{hf} values seem to follow a second order transition. However, in all cases at the reduced temperature values in the range of 0.5–0.6 (shown as two vertical dotted lines in Fig. 4b) there is an accentuated deviation from

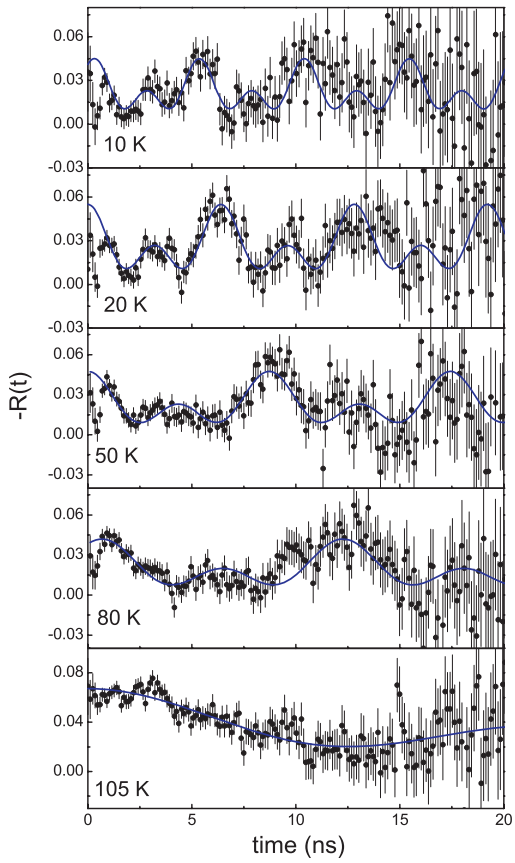

Fig. 3. Perturbation functions $R(t)$ of the ^{140}Ce probe in $GdRh_2Si_2$ at some temperatures below Néel temperature.

the behavior described by the Brillouin function. This deviation is ascribed to the polarization of localized moment at Ce ions induced by the magnetic field produced by the host moments [23] and can be quantitatively described by a model based on the molecular-field theory which considers that the localized moment on Ce ion is oriented by the exchange field of rare-earth ions of the host

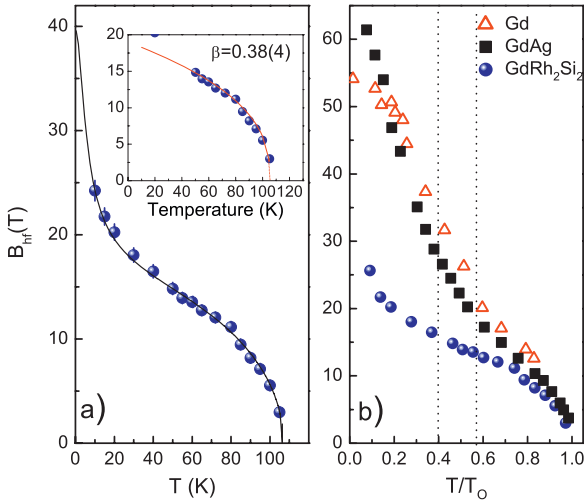

Fig. 4. (a) Temperature dependence of the magnetic hyperfine field at Gd sites occupied by ^{140}Ce probe in $GdRh_2Si_2$. The inset shows the behavior near the transition temperature. (b) Magnetic hyperfine field at Gd sites occupied by ^{140}Ce probe in Gd, GdAg, and $GdRh_2Si_2$ as a function of the reduced temperature T/T_0, where T_0 is the ordering temperature. Gd orders ferromagnetically at $T_0 = T_C = 292.2$ K [8], and GdAg orders antiferromagnetically at $T_0 = T_N = 132$ K [13].

Table 1
Parameters obtained from the fit of the model described in the text to the data of $GdRh_2Si_2$ and Gd, compared with those obtained for GdAg [12].

Compound	T_N (K)	$B^i_{hf}(0)$ (T)	$B^h_{hf}(0)$ (T)	ξ
$GdRh_2Si_2$	107	−28	12.4	0.056
GdAg	132	−65	7.5	0.148
Gd	292.2	−42.5	12.5	0.245

[11]. In this model the effective hyperfine field at the probe site is given by $B_{hf} = B^i_{hf} + B^h_{hf}$, where B^i_{hf} is the contribution from the probe ion itself, and B^h_{hf} is the contribution from the polarization of conduction electrons by the magnetic field of rare-earth ions of the host, which scales with the host reduced magnetization. B^i_{hf} is proportional to the thermal average of the impurity magnetic moment $\langle J^i \rangle$, which is localized. Therefore, the effective magnetic hyperfine field is $B_{hf}(T) = B^i_{hf}(0) \times B_{J^i}(y) + B^h_{hf}(0)\sigma(T)$, where $B_{J^i}(y)$ is the Brillouin function and y is its argument given by: $y = \mu_B(g_{J^i} - 1)\vec{J^i} \cdot \vec{B}^i_{exc}T)/kT$. In this expression, $B^i_{exc}(T) = B^i_{exc}(0)\xi\sigma(T)$, with $B^i_{exc}(0) = \{3kT_0/[(g_{J^h} - 1)(J^h + 1)\mu_B]\} \times \sigma(T)$ where T_0 is the magnetic transition temperature. The parameter ξ takes into account the fact that the host–impurity exchange may be different from the host–host exchange. The parameters $g_{J^h} = 2$ and $J^h = 7/2$ are, respectively, the Landè factor and total angular momentum of Gd ion. In Fig. 4a the solid curve represents the function described by this model which better fitted to the experimental data, where we have used $g_{J^i} = 6/7$ for the Landé factor and $J^i = 5/2$ as the total angular momentum of Ce^{3+}. Table 1 lists the relevant parameters obtained from the fit of the model to the experimental data. In this table, results for GdAg and Gd are also included. Data for GdAg were extracted from ref. [12] and data for Gd were obtained by applying the model to experimental data measured by Thiel et al. [8] The ξ values presented in Table 1 were normalized to $|[(g_{J^h} - 1)J^h]/[(g_{J^i} - 1)J^i]|$.

The contribution from the host magnetic ions to the mhf, $B^h_{hf}(0)$ comes from the polarization of conduction electrons induced by the local 4f magnetic moments of Gd ions of the intermetallic matrix. In general, as 4f-electrons are well localized, the magnetic coupling between the 4f-electrons in two nearest neighbor rare-earth ions occurs, therefore, by indirect exchange, which can be described by the RKKY theory based on the work of Ruderman and Kittel [24], Kasuya [25], and Yoshida [26] in which exchange interactions between magnetic ions in metals is mediated by conduction electrons which are spin-polarized by the localized 4f electrons in the rare-earth ions. Alternatively the indirect 4f–4f exchange can be also explained as being mediated by an intra-atomic 4f–5d polarization followed by the exchange interaction between the spin polarized 5d-electrons of neighboring rare-earth ions [27,28]. In both cases, a spin polarization arises which leads, via Fermi contact interaction, to a magnetic hyperfine field B_{hf} at the nucleus of a probe atom. In the RKKY interaction, polarized conduction electrons induce a non-zero spin density at the probe nuclei. On the other hand 5d electrons of ^{140}Ce probes are polarized via 4f–5d interaction, producing the magnetic hyperfine field. In both scenarios the mhf at a probe nucleus substituting one Gd ion is directly proportional to the number of magnetic nearest-neighbor atoms (N_{1nn}) and inversely proportional to the cube of the 1nn–1nn distance (d_{1nn}): $B^h_{hf}(0) \propto N_{1nn}/d^3_{1nn}$. Gd sites in $GdRh_2Si_2$ are surrounded by four Gd nearest neighbors in the a–b plane at a distance of 4.043 Å, as shown in Fig. 5b. Gd sites in GdAg have six Gd nearest neighbors: four of the Gd atoms with spin down in the ab plane, and two of them with spin up along the c-axis, all at a distance of 3.646 Å(see Fig. 5a). The Gd sites in pure Gd are surrounded by other twelve Gd at a distance of 3.64 Å. Considering that ^{140}Ce probes substitute Gd atoms in pure Gd, GdAg and $GdRh_2Si_2$ it is possible to compare the ratio $B^h_{hf}(0)d^3_{1nn}/N_{1nn}$ for each compound. The resulting ratios are 204.4 T Å^3, 182.4 T Å^3 and 50.4 T Å^3, respectively, for $GdRh_2Si_2$, GdAg and pure Gd. The value for Gd is much smaller as compared with the values for $GdRh_2Si_2$ and GdAg which are close to each other. These results indicate that the coupling mechanism between magnetic Gd ions in $GdRh_2Si_2$ and GdAg is similar, however different in pure Gd. If the mechanism of polarization in GdAg and $GdRh_2Si_2$ includes the conduction electrons, one possible explanation could be related to the contribution of Ag, and Rh and Si to the conduction electron density in GdAg and $GdRh_2Si_2$. This extra contribution seem to be decisively very important to the spin coupling between magnetic ions in GdAg and $GdRh_2Si_2$ compounds. The higher the conduction electron density, the more efficient the magnetic coupling is expected to be. Therefore, the density of conduction electrons in $GdRh_2Si_2$ must be higher, due to contribution from both Rh and Si, than in GdAg, which has only the contribution from Ag to the conduction electron density.

The results obtained from the fit also show that the $B^i_{hf}(0)$ value is much higher than $B^h_{hf}(0)$ and have opposite signs for all compound. This contribution to the magnetic hyperfine field is due to the polarization of f-spin moment of ^{140}Ce probe induced by the magnetic field coming from rare-earth moments of the matrix [23]. As $B^i_{hf}(0)$ for $GdRh_2Si_2$, GdAg and Gd is much smaller than the magnetic hyperfine field of 183 T for the free Ce^{3+} ion [29], it is assumed that the ground state for Ce impurity in those intermetallic compounds presents a valence state slightly higher than 3. Leal and

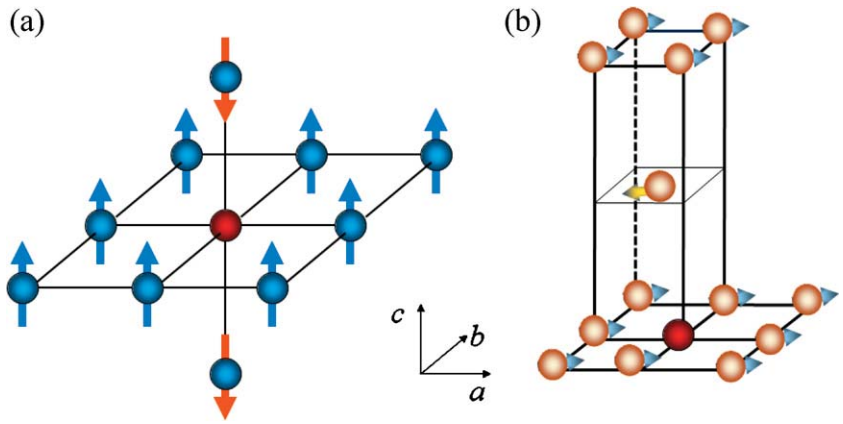

Fig. 5. Magnetic structure of (a) GdAg and (b) $GdRh_2Si_2$. Solid circles with arrows represent Gd atoms and their spin direction. Solid red circles without arrow represent ^{140}Ce probes. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

Troper [30], using an extension of the Daniel–Friedel model, performed calculations of the mhf at ^{140}Ce in pure Gd and obtained the value of $B_{hf} = -53.7$ T at $T = 0$ K. This value agrees very well with the value of 54 T measured by Thiel et al. [8] at 4.2 K. From these calculations the valence of Ce was estimated to be 3.25, rather than the usual 3 for Ce ions. As the mhf is even smaller in $GdRh_2Si_2$, it is reasonable to suppose that the valence of Ce ions in this compound is higher than that in pure Gd.

The magnetic interaction between a magnetic impurity ion diluted in a magnetic host is still an issue not well understood. The host–impurity exchange interaction, in particular, is not well described so far. In the model described above the strength of this interaction is measured by the parameter ξ. The higher the ξ value the weaker the interaction is. As shown in Table 1, this parameter is small for $GdRh_2Si_2$ when compared with the values for GdAg and pure Gd, indicating that the host–impurity exchange interaction is stronger in $GdRh_2Si_2$ than in GdAg and Gd, being the smallest in the latter case. As discussed above, the conduction electron density is higher in $GdRh_2Si_2$ than in GdAg, which it is probably higher than in Gd. Hence, one can straightforwardly infer that conduction electron density also plays an important role in this type of interaction. However, additional studies are needed in order to get a clearer understanding of this interaction.

4. Conclusions

The magnetic properties of $GdRh_2Si_2$ compound were investigated by the measurement of magnetic susceptibility and hyperfine magnetic field which provide a macroscopic and an atomic view of the magnetism, respectively. While the magnetic susceptibility shows a characteristic antiferromagnetic behavior as a function of temperature, the temperature dependence of mhf, measured with ^{140}Ce probes, shows an anomalous behavior with a sharp deviation from the expected curve described by the standard Brillouin function for an antiferromagnet. This unusual variation with temperature is due to an additional contribution to the mhf coming from the localized moment at Ce probe substituting Gd atoms, which is polarized by the exchange field of the antiferromagnetic host. With the use of a model based on the molecular field theory it was possible to calculate the contribution from the host and from the probe atom itself to the mhf. The comparison of the host contribution to the mhf of $GdRh_2Si_2$ with those of GdAg and pure Gd makes it possible to conclude that the contribution of Ag, and Rh and Si to the conduction electron density in $GdRh_2Si_2$ and GdAg compounds is certainly important to the spin coupling between magnetic ions of the host matrix in these compounds.

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