

Magnetic hyperfine interaction for Sn impurity atoms in RAI_2 ferromagnetic intermetallides: Spin and orbit contributions to the hyperfine field

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(Submitted 25 July 1984)

Zh. Eksp. Teor. Fiz. **88**, 300–308 (January 1985)

Mössbauer spectroscopy has been used to analyze the hyperfine interaction for ^{119}Sn impurity atoms replacing Al atoms in the ferromagnetic intermetallides RAI_2 , where $R = \text{Pr, Nd, Sm, Tb, Dy, Ho}$. The magnetic hyperfine fields, the quadrupole interaction constants, and the isomer shifts have been measured at $T = 5 \text{ K}$, and the dependence of the hyperfine field on the spin of the rare-earth (RE) ion has been studied. The dependences differ markedly for RAI_2 with light ($R = \text{Pr, Nd, Sm}$) and heavy RE ions ($R = \text{Tb, Dy, Ho}$), which indicates that for Sn impurity atoms, the orbital contribution to the magnetic hyperfine interaction is substantial. The orbital contribution adds to the spin contribution in intermetallides with light RE ions but subtracts from it in intermetallides with heavy RE ions. The experimental data can be represented as a linear superposition $B = aS_z + bD_z$ of the spin and orbit contributions, where B is the magnetic hyperfine field, S_z and L_z are the projections of the spin and orbital moments of the RE ion, $a = -4.16 \pm 0.14 \text{ T}/\mu_B$, $b = 1.05 \pm 0.08 \text{ T}/\mu_B$. The results qualitatively confirm the theoretical model proposed in Ref. 4 for transfer of orbital momentum to the nonmagnetic atom.

1. INTRODUCTION

Experimental data indicate that to first order, the magnetic hyperfine field B for nonmagnetic atoms in rare-earth (RE) magnets is proportional to the projection S_z of the RE ion spin on the z axis (cf. e.g. Refs. 1–3). This proportionality is usually regarded as a consequence of the proportionality between S_z and the polarization of the conduction electrons; however, there are other mechanisms for electron polarization transfer from the RE ion to the nonmagnetic atom for which B should also depend linearly on S_z .

The finding that B is not always strictly proportional to S_z is more unexpected and is difficult to explain. The departure from proportionality may be very great in many cases.³ The factors responsible for this nonlinearity may be important in the theory of the magnetic hyperfine interaction in RE magnets (and possibly also in the theory of the exchange interaction). Although several interpretations of the nonlinear dependence $B = B(S_z)$ have been discussed,^{3–6} none of them is convincingly supported by the experimental results.

The conjecture in Ref. 4, according to which the electron polarization contains an orbital contribution, is of special interest. A general analysis of the exchange interaction between the localized $4f$ -moment of the RE ion and the collectivized electrons implies that such a polarization can occur for the conduction electrons.⁴ Unfortunately, the magnitude of the orbital contribution to the conduction electron polarization is not amenable to theoretical estimate. Furthermore, the role of the conduction electrons in the hyperfine interaction remains imperfectly understood for nonmagnetic atoms. For example, the local nature of the hyperfine interaction noted in Ref. 7 for Sn impurity atoms in gadolinium intermetallides appears to contradict the usual assumption that the conduction electrons give a dominant contribution to the magnetic hyperfine field. A semiempirical formula was derived in Ref. 4 which includes both the spin and the orbit contributions to the hyperfine field. In

many ways, this formula accurately approximates the experimental data; however, it is difficult to interpret the empirical parameters found in this approximation.³ We note that almost all of the experimental data pertain to systems with heavy RE ions. Nevertheless, the amount of experimental material for each case is very scanty, and one cannot regard a successful approximation by a three-parameter formula as a confirmation of the theoretical model. Opinions vary as to whether orbital polarization can be transferred from an RE ion to a nonmagnetic atom. For instance, Bertier *et al.*⁵ assume that the orbital polarization is localized near the RE ion and has no influence on the B field at the neighboring nonmagnetic atoms.

In our present work we have attempted to obtain new experimental data aimed at determining how the orbital moment of the RE ion influences the magnetic hyperfine interaction for nonmagnetic atoms in RE ferromagnets. To this end we studied the hyperfine interaction for ^{119}Sn impurity atoms in RAI_2 ferromagnetic intermetallides containing both heavy ($R = \text{Tb, Dy, Ho}$) and light RE ions ($R = \text{Pr, Nd, Sm}$). (We previously obtained data for $R = \text{Gd}$ in Ref. 8.) The simplest and most straightforward way to check whether the orbital moment contributes to the hyperfine interaction is to compare the data for systems with light and heavy RE ions. The coupling scheme for the spin and orbital moments S, L differs from light and heavy RE ions: the moments are parallel for heavy RE ions ($L + S$ coupling) but antiparallel for light RE ions ($L - S$ coupling). Thus if the orbital contribution to B for one of these groups is added to the spin contribution, it will be subtracted from it for the other group. If the orbital contribution to B is nonzero and not too small, we expect that the dependences $B(S_z)$ will differ for the nonmagnetic atom in intermetallides with light and heavy RE ions. Our results indicate that this difference is indeed observable and thus lend qualitative support to the orbital polarization model suggested in Ref. 4.

2. EXPERIMENTAL METHOD AND PRINCIPAL RESULTS

We used Mössbauer spectroscopy to analyze the hyperfine interaction for ^{119}Sn impurity atoms in the intermetallics RAI_2 . It was shown previously^{7,8} that tin can form stable solid solutions in rare-earth intermetallics. The magnetic fields acting on the Sn nuclei are extremely strong, so that the absorption spectra have a well-resolved hyperfine structure.

2.1 Preparation of the alloys

The RAI_2 alloys were prepared by fusing the components (99.999% pure aluminum and 99.9% pure rare-earths) in an arc furnace in an argon atmosphere. The aluminum was first doped with metallic tin, which was enriched to 92% in the ^{119}Sn isotope. The concentration of the tin impurity was 0.5–0.6 at.%, and the alloys were annealed for 72 h at 800 °C. In most cases the annealing narrowed the resonance lines and improved the quality of the spectrum.

As in the case of gadolinium intermetallics,^{7,8} a small fraction of the tin did not form a solid solution, so that a spurious line was present near the center of the spectrum (perhaps some of the tin reacted directly with the rare earth during the melting process to form intermetallics of unknown composition). In any case, the spurious line became weaker during the annealing process. In some of the samples, this line became broader or split at low temperatures (usually below 10–20 K). Because the spurious line was weak and localized in the central region of the spectrum, and because the hyperfine structure of the primary spectrum was well-resolved, the presence of the extraneous phase did not cause serious difficulties in analyzing the absorption spectra.

2.2 Spectral measurements and analysis

The Mössbauer absorption spectra were recorded using a spectrometer with a constant acceleration. A resonant CaSnO_3 detector was employed to increase the resolution in detecting the Mössbauer radiation. All the measurements were made with a CaSnO_3 source at room temperature. The absorption spectra were recorded in external magnetic fields ≤ 0.9 T in order to determine the sign of the hyperfine field.

The measurements for all of the alloys were made for T from 5 K up to temperatures above the Curie point. In all cases the Curie temperatures, which were found from the temperature dependences of the hyperfine field, agreed closely with the data from the magnetic measurements in Ref. 9. In this article we will consider only the results found for $T = 5$ K.

The spectra were analyzed by the standard procedure of approximating them as a superposition of Lorentz lines. The spectral structure was well-resolved at $T = 5$ K, and we were able to determine the parameters of the hyperfine interaction without making any *a priori* model assumptions. As in the case of the GdAl_2 intermetallic,⁸ the analytical results imply unequivocally that the Sn impurity atoms must be localized at the Al-sites, since otherwise there would be no quadrupole shift in the lines and no anisotropic contribution to the hyperfine field.

2.3 Anisotropy of the magnetic hyperfine interaction. Quadrupole interaction and isomer shift

The RAI_2 intermetallics have the same $C15$ cubic structure as MgCu_2 ; the Mössbauer spectra for Sn impurity atoms in such matrices were considered in Ref. 8 for the case of GdAl_2 . The Al atoms are situated at the vertices of tetrahedra which center the octants of the cubic lattice of the RE atoms. All the Al sites are structurally equivalent and possess rhombohedral symmetry, with the third-order axis pointing along the [111] axes. The easy axis lies along one of the principal crystallographic planes: [100] for $R = \text{Pr, Nd, Dy}$, [110] for $R = \text{Ho}$, and [111] for $R = \text{Sm, Gd, Tb}$. Although the Al atoms are structurally equivalent, they are not equivalent in terms of the hyperfine interaction parameters because the angle θ between the local third-order axis and the magnetization vector differs for different lattice sites. The quadrupole shift in the components of the hyperfine structure and the anisotropic contribution to the magnetic hyperfine field are consequently not the same.

For magnetic moments pointing along the [111] axis, there are two groups of inequivalent Al-sites which are populated in the ratio 1:3 ($\theta = 0$ and $\theta = 70.5^\circ$). The quadrupole line shifts for these sites are equal to $e^2qQ/4$ and $-e^2qQ/12$, respectively (e^2qQ is the quadrupole interaction constant; q is the electric field gradient, and Q is the nuclear quadrupole moment). If the magnetic moments point along the [110] axis, there are again two groups of sites, which are now populated in the ratio 2:2 ($\theta = 35.3$ and 90°) and have the quadrupole shifts $e^2qQ/8$ and $-e^2qQ/8$. Finally, if the magnetic axis is parallel to the [100] crystallographic axis, all the Al sites are equivalent: $\theta = 54.7^\circ$, and the quadrupole shift therefore vanishes. Thus, if the magnetic moments are along the [111] or [110] axes, the spectrum should contain two sextets with the appropriate intensities, and quadrupole shifts should be observable; if the moments are along the [100] axis, there should be one sextet and no quadrupole shift. In all cases, our measured results for the directions of the easy axes coincide with published data. In particular, our results for HoAl_2 show unambiguously that the magnetic axis is directed along the [110] crystallographic axis, in agreement with recent magnetic measurements on single crystals in Ref. 10.

Figures 1 and 2 show some absorption spectra for ^{119}Sn in SmAl_2 and NdAl_2 ; Table I summarizes the fundamental results of the measurements. The sign of the hyperfine field was measured in the intermetallics NdAl_2 , SmAl_2 , and TbAl_2 . As anticipated, B is positive for the intermetallics with light RE ions and negative for intermetallics with heavy RE ions (and for GdAl_2 , cf. Ref. 8). This implies that B is always antiparallel to the spin of the RE ion. This finding is in agreement with the data for Sn in gadolinium intermetallics,⁷ and with data for other nonmagnetic atoms in RE ferromagnets (cf. e.g. Refs. 2, 6). The quadrupole shift is unobservable for the PrAl_2 , NdAl_2 , and DyAl_2 matrices in the ferromagnetic phase; in these cases we therefore measured e^2qQ at temperatures above the Curie point.

We see from Table I that the magnitudes of B differ substantially for Sn in SmAl_2 , GdAl_2 , TbAl_2 , and HoAl_2 ,

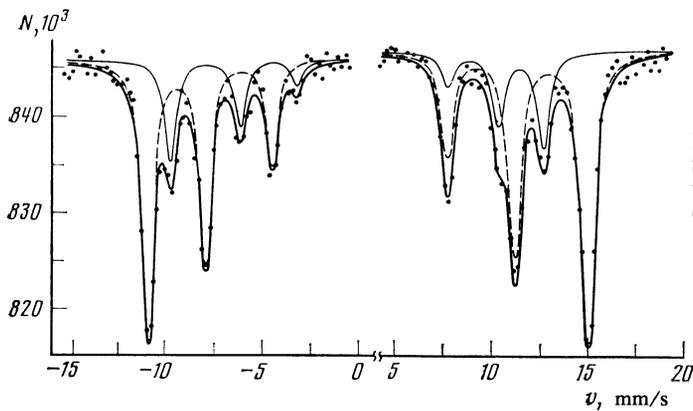
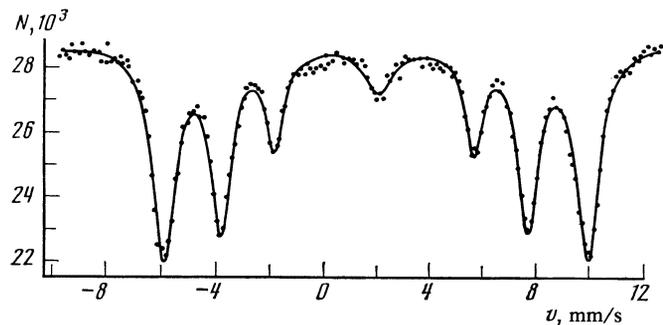


FIG. 1. Mössbauer absorption spectrum for ^{119}Sn impurity atoms in SmAl_2 recorded at $T = 5$ K. The thin continuous and dashed curves show the partial spectra corresponding to Sn atoms at sites with $\theta = 0$ and $\theta = 70.5^\circ$.

due to the anisotropic contribution to the hyperfine field. Two sources of anisotropic contribution may be considered: the dipole magnetic field from the localized moments of the RE ions,⁶ and the contribution caused by the anisotropic distribution of the electron density (both the density of the magnetic $4f$ -electrons and the density of the valence electrons of the RE ions and Sn atoms). The dipole field clearly cannot be entirely responsible for the anisotropic contribution to B for SmAl_2 and GdAl_2 : the sign of the anisotropic contribution in GdAl_2 is opposite to the sign of the dipole field,⁸ and the anisotropic contribution in SmAl_2 is ~ 20 times larger than the value calculated for the dipole field. We assumed in our discussion of the anisotropy of the hyperfine interaction in SmAl_2 in Ref. 11 that the anisotropic contribution to B reflects the nonsphericity of the distribution of the $4f$ -electrons of the Sm^{3+} ion. The RE ion in GdAl_2 is spherical; in this case, the anisotropic contribution may be attributed to the anisotropic polarization of the valence electrons of the Sn atom. Thus in all probability, the observed anisotropic contributions to B represent the sum of several contributions of different physical origin, some of which may partially cancel one another. Thus we conclude that the near-equality of the magnitudes of the anisotropic contribution and the dipole field for the Sn atoms in TbAl_2 and HoAl_2 is fortuitous.

In the subsequent discussion it will be convenient to consider only the isotropic part of the hyperfine field, which can be determined if we know the angular dependence of the anisotropic contribution. We assume that this dependence can be expressed in the form

$$B(\theta) = B_i + B_a(3 \cos^2 \theta - 1)/2, \quad (1)$$



where B_i and B_a are the isotropic and anisotropic components of the hyperfine field. Table I lists the isotropic components B_i calculated from Eq. (1). Since B_a is small compared to the total field B , the error in using (1) to find B_i is small and unimportant in the subsequent calculations.

The quadrupole interaction in SmAl_2 is also found to be anisotropic; for the other intermetallide matrices, the differences in the constants e^2qQ for the two types of sites is less than the measurement error. If we consider the average quadrupole interaction constants for SmAl_2 and TbAl_2 , we find that e^2qQ is almost constant and equal to 1.4–1.8 mm/s for the entire series of intermetallides. This implies that the chemical interactions between the Sn atoms and their neighboring atoms depend only weakly on the nature of the RE ion. The constancy of the isomer shift also indicates that the electronic configuration of the Sn atom is stable. Interestingly, the isomer shift for Sn in the intermetallides RAl_2 is almost equal to the shift for pure gadolinium¹² and for Gd intermetallides of various compositions.⁷ This is unexpected, since Al atoms rather than RE ions are the nearest neighbors of the Sn atoms in the RAl_2 matrices. Nevertheless, the magnitude of the isomer shift is characteristic of the Sn-RE interaction.

3. SPIN AND ORBIT CONTRIBUTIONS TO THE MAGNETIC HYPERFINE INTERACTION

In this section we will consider only the isotropic part B_i of the hyperfine field (for simplicity we will omit the subscript i in what follows).

As we have noted in the Introduction, we expect that to first order B will be proportional to the z -projection S_z of the

FIG. 2. Mössbauer absorption spectrum for ^{119}Sn impurity atoms in NdAl_2 recorded at $T = 5$ K. The line from the extraneous phase (cf. text) can be seen in the center of the spectrum.

TABLE I. Magnetic hyperfine fields B , quadrupole interaction constants e^2qQ , and isomer shifts IS for ^{119}Sn impurity atoms in RAl_2 ferromagnetic intermetallides.

R	θ	B , T	e^2qQ , mm/s	IS , mm/s	B_i , T
Pr	$54,7^\circ$	(+) 8,98 (2)	(-) 1,6 (1)	2,00 (2)	+8,98
Nd	$54,7^\circ$	+11,46 (2)	(-) 1,8 (1)	1,99 (2)	+11,46
Sm	0°	+16,32 (6)	-1,2 (2)	1,92 (4)	+18,3
	$70,5^\circ$	+18,94 (4)	-2,5 (4)	1,93 (2)	
Gd	0°	-28,6 (2)	-1,4 (2)	1,90 (3)	-29,2
	$70,5^\circ$	-29,3 (1)	-	1,90 (5)	
Tb	0°	-22,52 (9)	-2,3 (5)	2,03 (9)	-21,4
	$70,5^\circ$	-20,78 (4)	-1,3 (5)	1,92 (4)	
Dy	$54,7^\circ$	(-) 15,65 (9)	(-) 1,8 (1)	1,98 (8)	-15,65
Ho	$35,3^\circ$	(-) 12,2 (1)	(-) 1,4 (7)	1,93 (8)	-11,8
	90°	(-) 11,5 (1)	(-) 1,8 (6)	1,98 (8)	

Notes. 1) The isotropic components B_i of the hyperfine field were calculated using Eq. (1); θ is the angle between the local axis of third-order symmetry and the magnetic moment vector of the RE ions. 2) The + and - signs in the parentheses give the assumed signs of B and e^2qQ ; the constants e^2qQ were measured in the paramagnetic phase for the intermetallides with $R = \text{Pr}$, Nd , and Dy . 3) The data for Sn in GdAl_2 are taken from Ref. 8.

RE ion spin. We follow the usual procedure and assume first that S_z is equal to the projection of the spin in a free R^{3+} ion, i.e.,

$$S_z = J(g-1), \quad (2)$$

where J is the total momentum and g is the g -factor for the free ion. Since the Gd^{3+} ion has a pure spin moment ($S = 7/2$), it is convenient to normalize all fields B by the value of B for GdAl_2 . The expected dependence $B = B(S_z)$ can then be written in the form

$$B = J(g-1)B(\text{GdAl}_2)/3.5 = -8.32J(g-1)T. \quad (3)$$

The top part of Fig. 3 shows the experimental dependence $B(S_z)$ for a free ion. We see that B depends almost linearly on

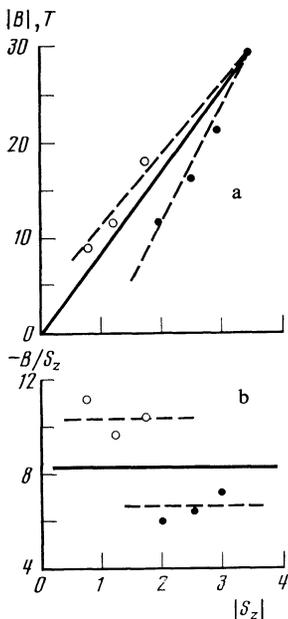


FIG. 3. Dependences of (a) the absolute magnitude B of the field and (b) $-B/S_z$ on $|S_z| = |J(g-1)|$. The solid line in (a) corresponds to Eq. (3). The horizontal solid line in (b) gives the ratio B/S_z for a zero orbital contribution to the hyperfine field. The light and dark circles give the values for the RAl_2 matrices for light ($R = \text{Pr}$, Nd , Sm) and heavy ($R = \text{Gd}$, Tb , Dy , Ho) RE ions, respectively. The dashed lines are shown for greater clarity.

S_z , i.e., the spin of the RE ion gives the main contribution to the hyperfine field, as expected. However, the experimental data clearly split cleanly into two groups corresponding to the intermetallides with light and heavy RE ions; these groups lie on either side of the straight line given by Eq. (3). The bottom part of Fig. 3, which plots B/S_z as a function of S_z , illustrates this division even more clearly. For all of the RAl_2 matrices with light RE ions, B/S_z is greater than for GdAl_2 , whereas the opposite is true for all the RAl_2 with heavy RE ions. Such a dichotomy is to be expected if an orbital contribution is present in addition to the spin contribution to B . We believe that the data in Fig. 3 convincingly demonstrate the existence of an additional orbital contribution to the hyperfine field, and that they lend qualitative support to the theoretical model proposed in Ref. 4.

Figure 3 shows that the sign of the orbital contribution to B is opposite to the sign of the spin contribution. For the light RE ions, for which L_z and S_z are antiparallel, both of these contributions are added and the observed field is stronger than is predicted by Eq. (3). For RAl_2 matrices with heavy RE ions L_z and S_z are parallel, so that the orbital contribution is subtracted from the spin contribution. The large magnitude of the orbital contribution is somewhat unexpected: according to the data in Fig. 3, the average orbital contribution comprises $\sim 20\%$ of the observed field B .

In order to analyze the results quantitatively we must discard the free-ion approximation and use the actual moments L_z , S_z for the RE ions in the intermetallides RAl_2 . The crystal field and exchange interaction effects are known to greatly influence the electronic configuration of the ions in many cases (and therefore also L_z and S_z). Unfortunately, the actual moments L_z and S_z in intermetallides are unknown, so that any quantitative analysis of the data must be only approximate. We will consider two methods for estimating L_z and S_z which are based on the experimental values for the magnetic moments of the ions in intermetallides: 1) If we assume that the crystal field primarily influences L_z but leaves S_z unchanged, we can find the projection L_z from the relation

$$M = L_z + 2S_z, \quad (4)$$

where M is the magnetic moment of the RE ion (divided by the Bohr magneton μ_B) and S_z is the projection of the spin for a free ion. 2) Alternatively, in the procedure suggested in Ref. 5 one assumes that the g -factor is constant (i.e., equal to its value for a free ion). We can then estimate J from the formula $M = gJ$ and find L_z and S_z . Both methods are only rough approximations. The first method is better if the crystal field effects are small, since in this case the use of S_z for a free ion cannot cause a large error. If the crystal field is not weak, the first method does not apply at all and the method proposed in Ref. 5 is to be preferred.

We note that the crystal field effects are not very strong in the intermetallides RAI_2 —in most cases the magnetic moment M of the ion differs by at most $(0.5\text{--}0.7)\mu_B$ from the value for a free ion. The magnetic hyperfine field for the RE field serves as a good test for how close L_z is to its value for a free ion, because the orbital moment gives the dominant contribution to this field. The experimental data show that for $SmAl_2$ (Ref. 13), $TbAl_2$ (Refs. 5, 13), and $DyAl_2$ (Ref. 5), the fields B for the R^{3+} ions are very close to B for the free ions, while for $PrAl_2$ and $NdAl_2$, B is substantially less than for the free ions.⁵ We therefore used the estimates in Ref. 5: $L_z = 3.76$, $S_z = -0.63$ and $L_z = 4.70$, $S_z = -0.74$ for $PrAl_2$ and $NdAl_2$, respectively. Elsewhere, we estimated L_z using Eq. (4): $L_z = 3.77, 0, 2.9, 4.8$, and 5.3 for $SmAl_2$, $GdAl_2$, $TbAl_2$, $DyAl_2$, and $HoAl_2$, respectively.

We chose the very simple formula

$$B = aS_z + bL_z, \quad (5)$$

or equivalently

$$B/S_z = a + b(L_z/S_z), \quad (6)$$

which expresses the experimental data as a linear superposition of the spin and orbital contributions. Figure 4 compares the results of (6) with the experimental data, and the agreement is seen to be very close. The following values of the coefficients a , b were found by a least-squares analysis: $a = -8.32 \pm 0.27$ T/spin unit $= -4.16 \pm 0.14$ T/ μ_B , $b = 1.05 \pm 0.08$ T/ μ_B . The value of a coincides with the ratio B/S_z for Sn in $GdAl_2$. We made no attempt to compare the experimental data with the three-parameter formula in Ref. 4 because the simpler formula (5) already describes the data accurately.

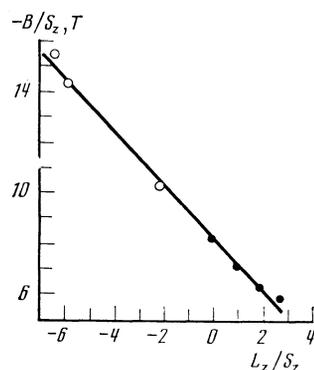


FIG. 4. Comparison of experimental data with Eq. (6) (cf. text). The solid line was found by a least squares analysis.

4. CONCLUSIONS

Our results show that both the spin and the orbital moments of the RE ion determine the magnetic hyperfine interaction for Sn impurity atoms in the intermetallides RAI_2 . The orbital contribution is not small and in principle can also be readily found for other systems. In particular, it would be interesting to study how the orbital contribution depends on the nature of the nonmagnetic atom. Unfortunately, most of the experimental data have been obtained for nonmagnetic impurity atoms in pure RE metals and are difficult to interpret, because the magnetic structure of the RE metals is diverse and occasionally very complicated. In addition, the hyperfine fields of nonmagnetic atoms have been measured almost exclusively in magnetic materials containing heavy RE ions. Reference 6 is an exception; NMR was employed there to measure B for ^{27}Al for the same intermetallides RAI_2 studied in the present paper, and we analyzed those results by the same method which we used to treat our data for Sn impurity atoms. We found that the dependences of B and B/S_z on S_z for Al have exactly the same form as the ones shown in Fig. 3 for Sn atoms. Moreover, Eq. (5) describes the data for Al just as accurately as the data for Sn impurity atoms. The coefficients in (5) for Al were found to be $a = -1.4 \pm 0.06$ T/spin unit $= -0.71 \pm 0.03$ T/ μ_B and $b = 0.24 \pm 0.02$ T/ μ_B . Because B is considerably less for Al than for Sn, the coefficients in Eq. (5) for Al are smaller. However, it is noteworthy that the ratios of the coefficients a and b for the two nonmagnetic atoms coincide to within the measurement error: $a(Al)/a(Sn) = 0.17 \pm 0.02$, $b(Al)/b(Sn) = 0.22 \pm 0.04$. This probably indicates that the same mechanism of electron polarization generates the magnetic field near the nucleus for Al and Sn impurity atoms.

Studies of the radial dependence of the orbital contribution to the hyperfine field will also be very useful for interpreting the results theoretically. For this purpose we can use the same method employed in Ref. 7, where the radial dependence of the spin contribution was studied for Sn impurity atoms in gadolinium intermetallides. One sees easily that the effects associated with the orbital contribution should be particularly pronounced in magnets containing RE ions at the beginning and end of the lanthanide series, since the orbital contribution to the hyperfine field may then be comparable to the spin contribution.

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