

# Magnetic hyperfine field for Sn-119 impurity atoms in gadolinium intermetallides

N. N. Delyagin, G. T. Mudzhiri, V. I. Nesterov, and S. I. Reiman

*Nuclear Physics Institute of the Moscow State University*

(Submitted 4 July 1983)

Zh. Eksp. Teor. Fiz. **86**, 1016–1025 (March 1984)

A Mössbauer-spectroscopy study is made of the magnetic hyperfine interaction for  $^{119}\text{Sn}$  impurity atoms in ferromagnetic gadolinium intermetallides. At a temperature of 5 K the magnetic hyperfine field  $B_{\text{hf}}$  is found to have the following values:  $-37.0 \pm 0.7$  T in  $\text{Gd}_3\text{In}$ ,  $-38.3 \pm 0.7$  T in  $\text{Gd}_2\text{In}$ , and  $(-32.4 \pm 0.6)$  T in  $\text{GdAg}_{0.5}\text{In}_{0.5}$ . The temperature dependence of  $B_{\text{hf}}$  is close to the Brillouin function for  $S = 7/2$ . For  $^{119}\text{Sn}$  in the intermetallide  $\text{Gd}_{0.95}\text{Y}_{0.05}\text{Al}_2$  a satellite structure appears in the spectrum as a result of the replacement of Gd by the nonmagnetic atoms Y. It can be inferred from analysis of this structure that in  $\text{GdAl}_2$  the Gd atoms of the closest coordination sphere are responsible for  $(90 \pm 10)\%$  of  $B_{\text{hf}}$ . The data on the magnetic hyperfine interaction for impurity Sn in gadolinium intermetallides and in gadolinium are interpreted in terms of a model in which the main contribution to  $B_{\text{hf}}$  in all of these systems is due to the nearest magnetic neighbors. The inferred radial dependence of the partial contributions to  $B_{\text{hf}}$  is compared with the analogous data for Sn in alloys of transition  $d$  metals. The conclusion is reached that in ferromagnets based on gadolinium there is no positive contribution to  $B_{\text{hf}}$  of the sort that competes in transition-metal alloys with the negative contribution from the nearest neighbors.

## 1. INTRODUCTION

We recently reported the results of the first study of the magnetic hyperfine interaction for Sn impurity atoms in a gadolinium intermetallide.<sup>1</sup> The magnetic hyperfine field ( $B_{\text{hf}}$ ) and its temperature dependence were measured by Mössbauer spectroscopy for  $^{119}\text{Sn}$  atoms which replace Al in the ferromagnetic intermetallic compound  $\text{GdAl}_2$ . The results of this work showed that tin forms a stable solid solution of replacement in  $\text{GdAl}_2$  and does not substantially influence the magnetic properties of the matrix. Further study along these lines using other rare earth intermetallides would seem extremely promising. The great diversity of structural and magnetic properties of rare earth intermetallides creates favorable conditions for investigating how the magnetic hyperfine interaction depends on the various parameters of the matrix. For checking theoretical models of the magnetic hyperfine interaction it is particularly interesting to study the radial dependence of the partial contributions to the hyperfine field, since this radial dependence contains more—complete and detailed information on the hyperfine—interaction mechanisms than does the total value  $B_{\text{hf}}$ . For Sn atoms in alloys of transition  $d$  metals the corresponding data were recently obtained<sup>2</sup> by comparing the value of  $B_{\text{hf}}$  in a large number of matrices having different radial distributions of the magnetic moments. One expects that a comparison of the radial dependences of the magnetic hyperfine interaction for magnets with substantially different electronic structures (transition  $d$  metals and rare earth elements) will prove extremely useful for putting certain basic assumptions of the theory of the magnetic hyperfine interaction on a more concrete footing. For example, one can expect to answer the question of the relative roles of the contact interaction with the conduction electrons and the direct overlap of the electronic wave functions. For choosing an adequate model of the magnetic hyperfine interaction this problem is of primary importance (see, e.g., Ref.

3), but its theoretical analysis is extremely complex and has still not produced unambiguous results.

In the present study we have investigated the magnetic hyperfine interaction for  $^{119}\text{Sn}$  impurity atoms in the ferromagnetic intermetallides  $\text{GdAg}_{0.5}\text{In}_{0.5}$ ,  $\text{Gd}_2\text{In}$ ,  $\text{Gd}_3\text{In}$ , and  $\text{Gd}_{0.95}\text{Y}_{0.05}\text{Al}_2$ . For Sn in the  $(\text{Gd}, \text{Y})\text{Al}_2$  matrix we have observed in the spectrum a satellite structure which can be analyzed to yield a direct determination of the contribution of the nearest magnetic neighbors in  $B_{\text{hf}}$ . We have also obtained some new data on the magnetic properties of the intermetallide  $\text{Gd}_2\text{In}$ .

## 2. MAGNETIC AND STRUCTURAL PROPERTIES OF THE INTERMETALLIDES $\text{CdAg}_{0.5}\text{In}_{0.5}$ , $\text{Gd}_2\text{In}$ , AND $\text{Gd}_3\text{In}$

The properties of the ordered alloys  $\text{GdAg}_x\text{In}_{1-x}$  have been studied in detail (see Refs. 4–6 and the literature cited therein). The compounds  $\text{GdAg}$  and  $\text{GdIn}$  are antiferromagnets, but their quasibinary solutions near  $x = 0.5$  are ferromagnetic. The Curie temperature ( $T_C$ ) is maximum for  $x = 0.5$  and is equal to 111–122 K,<sup>4,6</sup> and the magnetic moment of the Gd atoms is equal to  $7.3 \mu_B$ .<sup>5</sup> The alloy  $\text{GdAg}_{0.5}\text{In}_{0.5}$  has a cubic structure (of the CsCl type) with a lattice constant of 3.72 Å. The nearest magnetic neighbors of the Ag and In atoms are eight Gd atoms at a distance of 3.22 Å.

The data on the properties of the intermetallides  $\text{Gd}_2\text{In}$  and  $\text{Gd}_3\text{In}$  are extremely incomplete and contradictory. This is probably due in part to the high chemical activity of the alloys and to the difficulties in interpreting their behavior in external magnetic fields. In Ref. 7 the anomalies of the temperature dependence of the magnetic susceptibility of  $\text{Gd}_2\text{In}$  were attributed to a transition from an antiferromagnetic state to a ferromagnetic state at a temperature of 103 K; the value of  $T_C$  has not been determined. The results of our measurements (which will be discussed below) are consistent with this conclusion and show that the magnetic structure of  $\text{Gd}_2\text{In}$  remains ferromagnetic over the entire temperature

interval from 5 K to  $T_C$ . We note that the analogous magnetic anomalies observed in the intermetallides  $R_3Al_2$  were attributed not to a change in the spin configuration but to anomalies in the reaction of the domain structure to the external magnetic field.<sup>4,8</sup> One supposes that an analogous situation also occurs in the case of  $Gd_2In$  and that the "metamagnetic" behavior of this intermetallide in an external field does not reflect any anomalies of its magnetic structure at the microscopic level.

The intermetallide  $Gd_3In$  is a ferromagnet with  $T_C = 190\text{--}213$  K (Refs. 4, 9, 10). Magnetic saturation is not reached even in strong fields, and therefore the magnetic moment in the ferromagnetic state has not been measured.

The intermetallide  $Gd_2In$  has a structure of the  $Ni_2In$  type with lattice constants<sup>2</sup> of  $a = 5.413$  Å and  $c = 6.756$  Å. The nearest magnetic neighbors of the In atom are located at distances of 3.12 Å (three Gd atoms), 3.38 Å (two Gd atoms), and 3.55 Å (six Gd atoms). The crystal structure of  $Gd_3In$  has not been determined. The other rare earth compounds of the composition  $R_3In$  have either a face-centered cubic structure (of the  $Cu_3Au$  type) or its tetragonal modification ( $CuTi_3$  type). It can be assumed that  $Gd_3In$  also has one of these two structures. It is known that in the rare earth series the lattice constants for a given type of structure vary smoothly as a function of the atomic number of the rare earth element. Judging from this behavior and working in analogy with other rare earth compounds, we have estimated the probable values of the lattice constants of  $Gd_3In$  as  $a = 4.86$  Å for the  $Cu_3Au$  structure and  $a = 4.65$  Å and  $c = 4.99$  Å for the  $CuTi_3$  structure. The distance between nearest neighbors for both types of structure are approximately the same. We shall assume below that in  $Gd_3In$  the nearest neighbors of the In atom (the 12 Gd atoms) are located at a distance of 3.44 Å. The possible error of this value is probably not over 0.05 Å.

### 3. EXPERIMENTAL TECHNIQUES AND BASIC RESULTS

#### 1. Preparation of intermetallides and resonant absorbers

The compounds were prepared by the standard techniques (see the literature cited in the previous section). Metals of at least 99.9% purity were used. To increase the homogeneity, the ingots were remelted at least three times; the indium was preliminarily doped with tin enriched to 92% with the isotope  $^{119}Sn$ . Several alloys, containing from 0.3 to 1.0 at. %  $^{119}Sn$ , were prepared for each intermetallide; the results of the measurements were practically independent of the tin concentration (except for some broadening of the components of the hyperfine structure at the maximum concentrations). The main measurements were made with alloys containing around 0.5 at. %  $^{119}Sn$ .

Ingots of  $GdAg_{0.5}In_{0.5}$  were annealed in vacuum at 700 °C for 50 h. The intermetallides  $Gd_2In$  and  $Gd_3In$  formed in a well-ordered state directly from the melt, and so in these cases no additional heat treatment was done. (One of the samples of the alloy  $Gd_3In$  was annealed at 750 °C for 55 h; the annealing was found to have no appreciable affect on the Mössbauer spectrum.)

The intermetallide  $Gd_{0.95}Y_{0.05}Al_2$  was prepared by the

method used for  $GdAl_2$  in Ref. 1. The ingot was annealed at 750 °C for 55 h. The yttrium concentration corresponded to the replacement of 5.5% of the Gd atoms by the nonmagnetic atom Y.

The high chemical activity of intermetallides of the Gd-In system requires that careful precautions be taken, especially in working with the powders required for preparation of Mössbauer absorbers. Even a brief contact of the powders with air caused a marked degradation in sample quality. An isolated line (or an unresolved doublet) appeared in the center of the Mössbauer spectrum, indicating the formation of extraneous phases due to oxidation of the alloy. Prolonged contact with air caused the components of the magnetic hyperfine structure to vanish almost completely from the  $^{119}Sn$  spectra. A similar degradation of the quality of the alloys was observed upon contact with alcohol and even with liquid paraffin. The powders were therefore prepared at liquid-nitrogen temperature; after preparation the powders were rapidly sealed and were stored in liquid nitrogen. This procedure permitted us to obtain Mössbauer spectra in which the parasitic central line was either entirely absent or of low intensity. The powders were generally not annealed. Only for the alloy  $Gd(Ag,In)$  did we use a brief (1–2 h) annealing at 500–600 °C, since in this case the quality of the spectra was noticeably improved thereby.

#### 2. Measurements and analysis of the spectra

The particulars of the measurement and processing techniques for the Mössbauer absorption spectra were discussed in our earlier papers.<sup>1,12</sup> The measurements were made in the temperature range from 5 K to  $T_C$ . [For  $(Gd,Y)Al_2$  the spectrum was measured only at 77 K]. A Mössbauer radiation source in the form of  $CaSnO_3$  was held at room temperature. In some cases a spectrometer with a  $CaSnO_3$  resonance detector and laser interferometer were used to calibrate the velocity scale. The temperature of the resonant absorber was held constant to within 0.1 K or better. To determine the sign of  $B_{hf}$  the absorption spectra were measured in an external magnetic field of 0.9 T at a temperature of 77 K. For the alloy  $Gd(Ag,In)$  the sign of  $B_{hf}$  was not determined: In this case we were unable to observe a shift of the hyperfine-structure components in excess of the statistical accuracy of the measurements.

In all cases the spectra exhibited a well-resolved magnetic hyperfine structure, and so the spectra were analyzed using the simple procedure of approximating the spectrum by the sum of Lorentzian lines. The quadrupole shift of the hyperfine-structure components did not exceed 0.3 mm/sec. The widths of the components at low temperatures were 1.3–2.0 mm/sec; the broadening of the lines indicates the presence of a small scatter in the values of  $B_{hf}$  ( $\Delta B/B < 3\%$ ). The Curie temperature was determined from the temperature dependence of the magnetic hyperfine splittings.

#### 3. Results of the measurements

The main results of the measurements are presented in Table I.

Figure 1 shows the Mössbauer absorption spectrum for

TABLE I. Magnetic hyperfine field  $B_{\text{hf}}$ , isomer shifts for  $^{119}\text{Sn}$  impurity atoms in gadolinium intermetal- lides, and the Curie temperatures of the compounds.

Intermetallide	$B_{\text{hf}}, T$	Isomer shift, mm/sec	$T_C, K$
$\text{GdAg}_{0.5}\text{In}_{0.5}$	$(-)32.4 \pm 0.6$	$2.06 \pm 0.03$	$115 \pm 1$
$\text{Gd}_2\text{In}$	$-38.3 \pm 0.7$	$1.95 \pm 0.06$	$236 \pm 1$
$\text{Gd}_3\text{In}$	$-37.0 \pm 0.7$	$1.93 \pm 0.06$	$211 \pm 2$

Note.  $B_{\text{hf}}$  and the isomer shifts were measured at 5 K. The isomer shift was measured relative to a  $\text{CaSnO}_3$  source held at room temperature; for Sn in the  $\text{Gd}(\text{Ag}, \text{In})$  matrix the sign of  $B_{\text{hf}}$  was not determined;  $T_C$  was determined from the temperature dependence of the magnetic hyperfine fields.

$^{119}\text{Sn}$  in  $\text{Gd}_3\text{In}$  at a measurement temperature of 77 K. The spectra for Sn in  $\text{Gd}(\text{Ag}, \text{In})$  and  $\text{Gd}_2\text{In}$  had an analogous form.

The temperature dependence of  $B_{\text{hf}}(T)$  is shown in Figs. 2 and 3. It is seen that the curves do not have features of any kind and are extremely close to the Brillouin functions for  $S = 1/2$ . In view of the high sensitivity of the hyperfine field for Sn to features of the spin structure of the matrix, one can conclude that for  $\text{GdAg}_{0.5}\text{In}_{0.5}$ ,  $\text{Gd}_2\text{In}$ , and  $\text{Gd}_3\text{In}$  this structure remains unchanged over the entire temperature range from 5 K to  $T_C$ . For  $\text{GdAg}_{0.5}\text{In}_{0.5}$  and  $\text{Gd}_3\text{In}$  the values of  $T_C$  found in this experiment are in good agreement with published data; for  $\text{Gd}_2\text{In}$  the value of  $T_C$  was determined here for the first time.

As can be seen from Table I, the isomer shift of the Mössbauer line is almost independent of the composition of the intermetallide. The same isomer shift was also obtained previously<sup>1</sup> for  $^{119}\text{Sn}$  in  $\text{GdAl}_2$ .

The spectrum for  $^{119}\text{Sn}$  in the  $\text{Gd}_{0.95}\text{Y}_{0.05}\text{Al}_2$  matrix was measured for the purpose of determining how  $B_{\text{hf}}$  changes when the magnetic atom Gd is replaced by the nonmagnetic atom Y. The replacement method is widely used to study the radial dependence of the magnetic hyperfine interaction. Under favorable conditions a satellite structure can be seen in the spectra; the shift of the hyperfine-structure component corresponds to the change in  $B_{\text{hf}}$  upon replacement of the magnetic atom by the nonmagnetic atom. Such a structure can actually be observed only in cases where  $B_{\text{hf}}$  is due to a small number of magnetic coordination spheres (or where the contribution of one of the coordination spheres is predominant). For Sn in alloys of transition  $d$  metals the contributions to  $B_{\text{hf}}$  from the nearest magnetic neighbors

and the magnetic atoms in the more remote spheres are comparable in absolute value,<sup>2</sup> and for this reason a resolved satellite structure which would admit and unambiguous interpretation is usually not observed.

The results obtained for Sn in  $(\text{Gd}, \text{Y})\text{Al}_2$  show that the situation in rare earth magnets is substantially different from that which obtains in magnetic alloys of transition metals. For Sn in  $(\text{Gd}, \text{Y})\text{Al}_2$  we observed a distinct satellite structure which is easily interpreted under the assumption that the shift of the hyperfine-structure components is due to the replacement of the Gd atoms by Y atoms in the first coordination sphere.

Figure 4 shows a part of the spectrum for  $^{119}\text{Sn}$  in  $(\text{Gd}, \text{Y})\text{Al}_2$  (the components located at negative velocities). In addition to the lines coinciding with components of the spectrum for  $^{119}\text{Sn}$  in  $\text{GdAl}_2$  (Ref. 1), satellite lines due to the replacement of a fraction of the Gd atoms by the nonmagnetic atoms Y appear in the spectrum. The position of the satellites is well determined, so that the spectrum can be analyzed (decomposed into the sum of partial spectra) without any important additional assumptions. In processing the spectrum we neglected the change in the dipole contribution to the magnetic hyperfine interaction, since, according to the calculations, the change in the dipole field upon the replacement Gd by Y is not over 0.25 T and the corresponding shift of the hyperfine-structure components is small in comparison with the linewidth. We also neglected the shift due to the anisotropic contribution to the magnetic hyperfine interaction and the shift due to the quadrupole interaction, since for the components shown in Fig. 4 these shifts are also small.<sup>1</sup>

The position of the first satellite between the two outside components of the main spectrum indicates that  $B_{\text{hf}}$  is

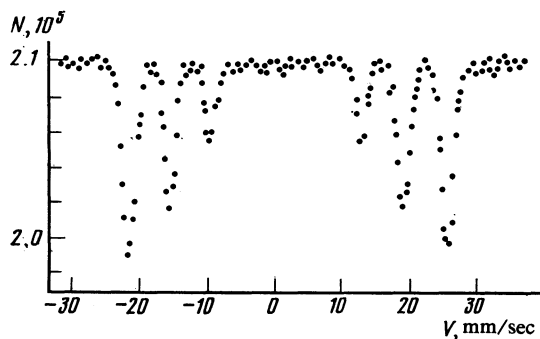


FIG. 1. Mössbauer spectrum measured at 77 K for  $^{119}\text{Sn}$  impurity atoms in  $\text{Gd}_3\text{In}$ .

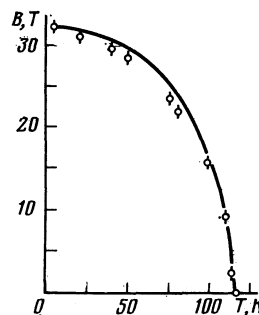


FIG. 2. Temperature dependence of the magnetic hyperfine field for  $^{119}\text{Sn}$  impurity atoms in  $\text{GdAg}_{0.5}\text{In}_{0.5}$ . The solid curve is the Brillouin function for  $S = 7/2$ .

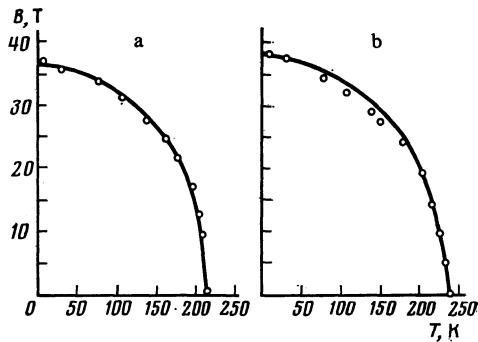


FIG. 3. Temperature dependence of the magnetic hyperfine field for  $^{119}\text{Sn}$  in  $\text{Gd}_3\text{In}$  (a) and  $\text{Gd}_2\text{In}$  (b). The solid curve is the Brillouin function for  $S = 7/2$ .

decreased strongly by the replacement of the Gd atom by Y. It is natural to suppose that the decrease in  $B_{\text{hf}}$  is due to the occurrence of the nonmagnetic atom Y in the closest coordination sphere. In  $\text{GdAl}_2$  this sphere contains six Gd atoms at a distance of 3.27 Å (Ref. 1). If the Y atoms are distributed statistically over the Gd sites, the probability for  $n$  yttrium atoms to occur in the first coordination sphere is

$$W(n) = C_0^n p^n q^n,$$

where  $p$  and  $q$  are the relative concentrations of gadolinium and yttrium. For our alloy ( $p = 0.945$ ,  $q = 0.055$ ) the configurations with  $n = 0, 1$ , and 2 should occur in the proportions  $W(0):W(1):W(2) = 1:0.34:0.04$ . Figure 4 shows the result of a decomposition of the spectrum into the three partial spectra which correspond to these three configurations. In the decomposition procedure the shifts, widths, and intensities of the components were treated as free parameters. As can be seen in Fig. 4, the sum of the resulting partial spectra approximates the experimental spectrum very well. The parameters found for the first partial spectrum ( $n = 0$ ) agree with the parameters<sup>1</sup> of the spectrum for Sn in  $\text{GdAl}_2$ . The areal ratio of the  $n = 0$  and  $n = 1$  spectra is  $0.35 \pm 0.02$ , in excellent agreement with the calculated ratio  $W(1)/W(0)$ . These results confirm the correctness of the model used to process the spectrum. The absolute value of  $B_{\text{hf}}$  for the configurations with  $n = 1$  and  $n = 2$  decreased in comparison with the value of  $B_{\text{hf}}$  for the  $n = 0$  configuration by  $3.3 \pm 0.5$  T and  $7.5 \pm 0.8$  T, respectively.

At a temperature of 77 K the hyperfine field for Sn in

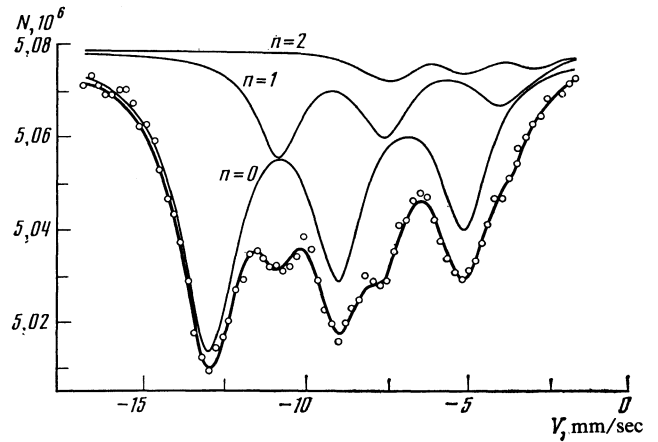


FIG. 4. Mössbauer spectrum measured at 77 K for  $^{119}\text{Sn}$  impurity atoms in  $\text{Gd}_{0.95}\text{Y}_{0.05}\text{Al}_2$ . The thin lines show the three partial spectra for configurations with  $n = 0, 1$ , and 2 yttrium atoms in the first coordination sphere. The thick curve at the bottom is the sum of the partial spectra.

$\text{GdAl}_2$  is  $-22.4$  T (Ref. 1). If  $B_{\text{hf}}$  is, on the whole, due to the magnetic atoms of the first coordination sphere, then the replacement of one or two Gd atoms by Y atoms should lead to a decrease in the absolute value of  $B_{\text{hf}}$  by 3.73 T or 7.47 T, respectively ( $1/6$  and  $2/6$  of the value of  $B_{\text{hf}}$  in  $\text{GdAl}_2$ ). The experimentally determined changes in the hyperfine field are in good agreement with these values. In view of the statistical error in the measurements, one can conclude that for the Sn atoms localized at Al sites in the  $\text{GdAl}_2$  matrix, the Gd atoms in the closest coordination sphere are responsible for  $(90 \pm 10)\%$  of the total value of  $B_{\text{hf}}$ .

#### 4. RADIAL DEPENDENCE OF THE CONTRIBUTIONS TO $B_{\text{hf}}$ FOR Sn ATOMS IN GADOLINIUM INTERMETALLIDES

On the basis of these results one can draw several conclusions about the radial dependence of the partial contributions to  $B_{\text{hf}}$  for Sn impurity atoms in gadolinium intermetallics. Table II gives all the currently known data on the values of  $B_{\text{hf}}$  for Sn atoms in metallic ferromagnets based on gadolinium. The magnetic hyperfine fields are negative, with absolute values lying in the narrow range 30–40 T. It is noteworthy that  $B_{\text{hf}}$  is relatively independent of the structure of the matrix and also of the relative sizes of the magnetic moment  $M_1$  of the first coordination sphere and the aver-

TABLE II. Magnetic hyperfine field  $B_{\text{hf}}$  for Sn impurity atoms and the partial contributions  $p(r_1)$  calculated from formula (1).

Matrix	$B_{\text{hf}}, \text{T}$	$\mu, \mu_B$	$r_1, \text{Å}$	$N$	$M_1, \mu_B$	$p(r_1), \text{T}/\mu_B$
Gd	-32.9 [13]	7.5	3.60	12	90.0	-0.37
$\text{GdAl}_2$	-29.3 [1]	7.1	3.27	6	42.6	-0.69
$\text{GdAg}_{0.5}\text{In}_{0.5}$	(-) 32.4	7.3	3.22	8	58.4	-0.55
$\text{Gd}_3\text{In}$	-37.0	(7.3)	(3.44)	12	87.6	-0.42
$\text{Gd}_2\text{In}$	-38.3	(7.3)	3.12	3	21.9	-0.66
			3.38	2	14.6	
			3.55	6	43.8	

Note.  $\mu$  is the magnetic moment of the Gd atom,  $N$  is the number of Gd atoms located at distance  $r_1$ ,  $M_1$  is the total magnetic moment of these atoms ( $M_1 = \mu N$ ), and the values in parentheses are conjectural (see main text).

age atomic magnetic moment of the alloy. This independence is in sharp contrast with the behavior found for Sn atoms in ferromagnetic alloys based on transition  $d$  metals.<sup>2</sup> In transition-metal alloys  $B_{\text{hf}}$  is determined by competing contributions of opposite signs and comparable absolute values: The contribution from the nearest magnetic neighbors is negative and the contribution from more remote magnetic atoms is positive. As a result,  $B_{\text{hf}}$  varies over wide limits, being sometimes positive and sometimes negative depending on the relative size of  $M_1$  and the average magnetic moment of the alloy. It seems clear that the radial dependence of the contributions to  $B_{\text{hf}}$  in the gadolinium intermetallides should be of a substantially different character.

To explain the experimental data, let us turn to the result obtained for the matrix  $(\text{Gd},\text{Y})\text{Al}_2$ . Let us assume that this result is of a general nature, i.e., that for all the matrices represented in Table II the dominant contribution to  $B_{\text{hf}}$  is from the Gd atoms of the first coordination sphere. In view of the small collection of data currently available, it is advisable to consider only the simplest model, assuming that  $B_{\text{hf}}$  is due entirely to the nearest neighbors (for all the matrices in Table II the distance to the second magnetic sphere is greater than 5 Å). It is also natural to suppose that in all the investigated intermetallides the Sn impurity atoms replace atoms of the nontransition element, i.e., Ag, Al, or In (for  $\text{GdAl}_2$ ) this follows directly from the experimental data<sup>1</sup>). In this model the general expression for  $B_{\text{hf}}$  in terms of the partial contributions<sup>2</sup> takes on the simple form

$$B_{\text{hf}} = M_1 p(r_1), \quad (1)$$

where  $p(r_1)$  is the partial contribution (in units of  $T/\mu_B$ ) to  $B_{\text{hf}}$  for a moment of  $1 \mu_B$  located at a distance  $r_1$  from the Sn atom ( $M_1$  is in units of  $\mu_B$ ). With the data of Table II one can use formula (1) to find the partial contributions  $p(r_1)$  which determine  $B_{\text{hf}}$  for Sn impurity atoms. For  $\text{Gd}_2\text{In}$  and  $\text{Gd}_3\text{In}$  the magnetic moments of the Gd atoms have not been determined. It is known,<sup>4</sup> however, that the moment of Gd in alloys and intermetallides generally lies in the range 7.0–7.5  $\mu_B$ ; as a reasonable estimate we have adopted a value of 7.3  $\mu_B$  for this moment in  $\text{Gd}_2\text{In}$  and  $\text{Gd}_3\text{In}$ . For the matrices Gd,  $\text{GdAl}_2$ ,  $\text{Gd}(\text{Ag},\text{In})$ , and  $\text{Gd}_3\text{In}$  the partial contributions  $p(r_1)$  are calculated directly with formula (1); the results of the calculations are given in the last column of Table II (in Gd six of the “nearest” neighbors lie at a distance of 3.572 Å and the other six at 3.636 Å; for simplicity we have neglected this small difference in  $r_1$ ). For the matrix  $\text{Gd}_2\text{In}$  one should consider three groups of “nearest” neighbors with different  $r_1$  (see Table II).

In this case formula (1) becomes

$$-38.3 = 21.9p(3.12) + 14.6p(3.38) + 43.8p(3.55). \quad (2)$$

Using the data obtained for different matrices and taking  $p(3.55) \approx p(3.60) = -0.37 T/\mu_B$  and  $p(3.38) = -0.52 T/\mu_B$  [obtained by linear interpolation between the values of  $p(3.44)$  and  $p(3.27)$ ], we find from (2) the value  $p(3.12) = -0.66 T/\mu_B$ .

The resulting radial dependence of the partial contributions  $p(r)$  is shown in Fig. 5. In view of the approximate nature of the model and of the assumptions made, the model

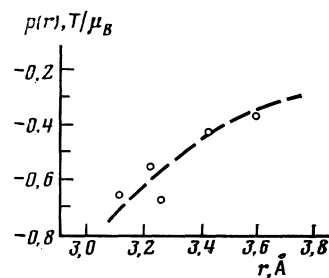


FIG. 5. Radial dependence of the partial contributions  $p(r)$  to the magnetic hyperfine field for tin impurity atoms in gadolinium and in gadolinium intermetallides. The partial contributions were calculated with formulas (1) and (2). The dashed curve shows the hypothetical form of the function  $p(r)$ .

should be regarded as giving a consistent and reasonable explanation for the experimental data. On the average, the partial contributions fall off in absolute value as the distance increases, as one would expect of polarization mechanisms for the nearest-neighbor interactions. As in the case of transition-metal alloys, the nearest magnetic neighbors give a negative contribution to the hyperfine field, but the positive contribution from more remote neighbors is absent (or extremely small). It is for this reason that  $B_{\text{hf}}$  is comparatively large in absolute value for Sn atoms in gadolinium intermetallides in spite of the relatively small values of  $p(r)$ . In ferromagnets based on  $d$  metals the partial contributions for the first coordination sphere are much larger, between  $-2$  and  $-3 T/\mu_B$  (Ref. 2), but the corresponding negative contribution is compensated to varying degrees by a positive contribution of comparable size. In gadolinium intermetallides there is no such compensation. Subsequent experiments should reveal to what extent this conclusion is of a general nature. In this regard we should mention the paper by Eckrich *et al.*,<sup>14</sup> who showed by analyzing the NMR spectra for  $^{67}\text{Zn}$  that around 80% of  $B_{\text{hf}}$  for Zn in the intermetallides  $\text{RZn}$  is due to the nearest magnetic neighbors.

The above results show that in gadolinium intermetallides the electronic-polarization mechanisms responsible for  $B_{\text{hf}}$  have a comparatively small effective radius. The conclusion that the magnetic hyperfine interaction for Sn atoms in rare earth intermetallides is relatively “localized” is extremely unexpected. The models currently used to describe the magnetic hyperfine interaction for nonmagnetic atoms in metallic magnets usually consider two possible sources of the hyperfine field: the contact interaction with polarized conduction electrons, and the overlap of the electrons of the nonmagnetic atom with the magnetic electrons of the neighboring atoms of the matrix. For magnets based on rare earth elements the second of these two mechanisms is usually ignored, since the magnetic 4f electrons are well localized and screened by the outer-shell electrons. The interaction with conduction electrons is traditionally treated as an interaction with a large effective radius, since the polarization of the electrons on the Fermi surface is a relatively weak function of the distance. On the basis of these ideas it is hard to explain why the contribution determined by the interaction at large distances should vanish in the gadolinium intermetallides (as opposed to alloys of transition  $d$  metals). This diffi-

culty can be eliminated by assuming that the large-radius interaction in transition-metal alloys is due not to the  $s$  conduction electrons but to the collectivized magnetic 3d electrons (which are absent in the gadolinium intermetallides). Such an assumption, however, would require a substantial revision of the traditional ideas and can scarcely be justified without further experimental and theoretical studies.

From an experimental standpoint it is interesting to compare the derivatives  $\partial p/\partial r$  for Sn atoms in d-metal alloys and gadolinium intermetallides. In d-metal magnets the negative contribution to  $B_{\text{hf}}$  is characterized by a very strong dependence on distance. For  $r = 2.45\text{--}2.60 \text{ \AA}$  (typical nearest-neighbor distances in transition-metal alloys) the derivatives  $\partial p/\partial r$  are equal to  $10\text{--}14 T/\mu_B \text{ \AA}$  (Refs. 2, 15). For this reason, effects due to the radial dependence of the magnetic hyperfine interaction (a strong pressure dependence<sup>16</sup> of  $B_{\text{hf}}$  and the presence of temperature anomalies due to the thermal expansion<sup>15,17</sup>) are clearly manifested for Sn in transition-metal alloys. In gadolinium intermetallides the derivative  $\partial p/\partial r$  is much smaller; it can be estimated from the data of Fig. 5 as  $0.5\text{--}1.0 T/\mu_B \text{ \AA}$ . Thus for Sn in rare earth alloys one should not expect the large temperature anomalies in  $B_{\text{hf}}$  that are so characteristic for Sn in d-metal alloys. It might also be noted that in alloys whose only magnetic atoms are Gd, one should not observe positive hyperfine fields for Sn. A different situation can arise in intermetallides which, in addition to Gd atoms, also contain 3d metals having a nonzero magnetic moment. The study of the mag-

netic hyperfine interaction in these alloys would permit comparison of the contributions to  $B_{\text{hf}}$  for the 3d and 4f atoms in systems having the same electronic structure.

- <sup>1</sup>N. N. Delyagin, V. I. Nesterov, and S. I. Reĭman, Zh. Eksp. Teor. Fiz. **84**, 1580 (1983) [Sov. Phys. JETP **57**, 922 (1983)].
- <sup>2</sup>N. N. Delyagin, V. I. Drylov, and V. I. Nesterov, Zh. Eksp. Teor. Fiz. **79**, 1049 (1980) [Sov. Phys. JETP **52**, 532 (1980)].
- <sup>3</sup>M. B. Stearns and J. M. Norbeck, Phys. Rev. B **20**, 3739 (1979).
- <sup>4</sup>K. H. J. Buschow, Rep. Prog. Phys. **42**, 1373 (1979).
- <sup>5</sup>K. H. J. Buschow, A. Oppelt, and E. Dormann, Phys. Status Solidi B **50**, 647 (1972).
- <sup>6</sup>K. Yagasaki, Y. Hidaka, H. Fujii, and T. Okamoto, J. Phys. Soc. Jpn. **45**, 110 (1978).
- <sup>7</sup>H. Gamari-Seale, T. Anagnostopoulos, and J. R. Yakinthos, J. Appl. Phys. **50**, 434 (1979).
- <sup>8</sup>K. N. R. Taylor, Adv. Phys. **20**, 551 (1971).
- <sup>9</sup>R. D. Hutchens and W. E. Wallace, J. Solid State Chem. **9**, 152 (1974).
- <sup>10</sup>S. Parviainen, Phys. Status Solidi A **60**, K13 (1980); S. Parviainen and S. Penttila, Phys. Status Solidi A **60**, K119 (1980).
- <sup>11</sup>A. Palenzona, J. Less-Common Met. **16**, 379 (1968).
- <sup>12</sup>V. I. Krylov, V. P. Gor'kov, N. N. Delyagin, Yu. D. Zonnenberg, and V. I. Nesterov, Zh. Eksp. Teor. Fiz. **77**, 2093 (1979) [Sov. Phys. JETP **50**, 1000 (1979)].
- <sup>13</sup>V. Gotthardt, H. S. Moller, and R. L. Mössbauer, Phys. Lett. A **28**, 480 (1969).
- <sup>14</sup>K. Eckrich, E. Dormann, A. Oppelt, and K. H. J. Buschow, Z. Phys. B **23**, 157 (1976).
- <sup>15</sup>N. N. Delyagin, Yu. D. Zonnenberg, V. I. Krylov, and V. I. Nesterov, Hyperfine Interactions **11**, 65 (1981).
- <sup>16</sup>I. N. Nikolaev, V. P. Potapov, and N. N. Delyagin, Zh. Eksp. Teor. Fiz. **70**, 241 (1976) [Sov. Phys. JETP **43**, 125 (1976)].
- <sup>17</sup>N. N. Delyagin, V. I. Krylov, and V. I. Nesterov, Phys. Status Solid B **90**, 741 (1978); Solid State Commun. **32**, 805 (1979).

Translated by Steve Torstveit