Hyperfine interaction for ^{119}Sn impurity atoms in the ferromagnetic intermetallic compound GdAl$_2$

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The hyperfine interaction for ^{119}Sn impurity atoms in the ferromagnetic intermetallic compound GdAl$_2$ has been studied by Mössbauer spectroscopy in the temperature range from 6 to 300 K. The structure of the absorption spectra corresponds to localization of the Sn atoms on Al sites. The hyperfine field for Sn atoms at 6 K is $B_{hf} = -29.3 \pm 0.1T$. On increasing the temperature, the hyperfine field decreases somewhat faster than the magnetization of the matrix. The value found for B_{hf} for tin in GdAl$_2$ is close to the value for Sn in pure gadolinium, indicating a strong radial dependence of the partial contributions to the hyperfine field. As in alloys of transition d-metals, a decrease in the distance between the atoms is accompanied by an increase in the negative contribution to B_{hf}, Estimates are obtained for the electric field gradient in the vicinity of the Sn nucleus and for the anisotropic contribution to the magnetic hyperfine interaction.

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1. INTRODUCTION

The magnetic hyperfine interaction (MHFI) for nonmagnetic atoms in metallic materials is being studied intensively both experimentally and theoretically. In spite of the noticeable progress which has been achieved in this field recently, the interpretation of experimental results still meets with serious difficulties, mainly due to the lack of a correct theoretical model capable of explaining a wide range of experimental results from a single point of view. The main attention in theoretical work is paid to interpretation of the hyperfine fields for impurity atoms in homogeneous ferromagnetic matrices (Fe,Ni). However, these data can be explained within different (often diametrically opposed) concepts, and the question of the unique and correct choice of polarization mechanisms that produce the magnetic hyperfine field remains open.

Particular attention in this connection should be paid to studies of MHFI in systems with an inhomogeneous distribution of local magnetization, i.e., in ordered magnetic alloys and intermetallic compounds. Results of such studies contain information on the most important characteristic of MHFI, the radial distribution function $p(r)$ of the partial contributions to the magnetic hyperfine field.[1] This function, being a "differential" characteristic of the hyperfine interaction, contains fuller and more detailed information on the polarization mechanisms than the total value of the hyperfine field. Unfortunately, there are many difficulties in a direct measurement of the parameters of the function $p(r)$ and it is only possible in a limited number of cases. Indirect data on $p(r)$ can be obtained from measurement of hyperfine interaction in ordered systems with a different radial distribution of magnetic moments and with various interatomic distances. An empirical $p(r)$ function for Sn in metallic magnetic materials could be constructed by analysis of many measurements made for Sn atoms in magnetic alloys of transition d–metals. Results of these studies are summarized by Delyagin *et al.*[1]

It would undoubtedly be interesting to obtain similar results for Sn atoms in magnetically ordered alloys and intermetallic compounds of the rare earths. These results would be extremely valuable for interpreting some interesting results obtained in studying MHFI for Sn impurity atoms in pure rare earth metals.[2,3] A comparison of the characteristics of the $p(r)$ function in magnetic materials based on transition d-metals and in rare earths could be very significant for the development of the theory of MHFI. We have at present no information about the radial dependence of MHFI for Sn atoms in rare earth metals. So far, measurements of MHFI parameters for Sn impurity atoms by the method of Mössbauer spectroscopy have only been carried out for homogeneous metallic matrices.[2,3]

We report here the first successful attempt at studying hyperfine interaction for Sn impurity atoms in intermetallic gadolinium alloys. The first material studied was GdAl$_2$ which belongs to the extensive Laves-phase family. These compounds have a broad variety of magnetic properties which makes their study especially promising. We should point out that tin is one of the few elements which does not form a Laves phase. This raised some doubt about whether a stable solid solution of tin in GdAl$_2$ could be achieved. However, such a solution was obtained, which was very important for the further development of these studies.

2. STRUCTURE AND MAGNETIC PROPERTIES OF GdAl$_2$

The Laves phase GdAl$_2$ has C 15 cubic structure (of the MgCu$_2$ type) with lattice constant $a = 7.90$ Å. The Gd atoms form a face-centered-cubic lattice, the octants of which have alternately Gd atoms and right tetrahedra of Al atoms in the center. Two octants of the GdAl$_2$ elementary cell are shown in Fig. 1. All Gd atoms are structurally equivalent, as are the Al atoms. The Gd sites have cubic point symmetry, similar to the symmetry of atoms in the diamond lattice. The Al sites have rhombohedral symmetry with a threefold axis directed along one of the [111] axes.

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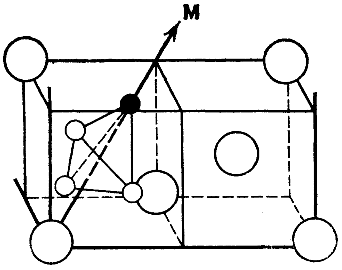

FIG. 1. Two adjacent octants of the $GdAl_2$ elementary cell. The arrow indicates the direction of the easy magnetization axis (the [111] axis); ○ (large)—Gd, ●—Al (*a* sites), ○—Al (*b* sites).

$GdAl_2$ is a collinear ferromagnet with Curie temperature $T_c \approx 170$ K. The magnetic moment per formula unit (i.e., the magnetic moment of a Gd atom) is $7.1 \pm 0.1\,\mu_B$.[4-8] NMR studies[9,10] of the hyperfine interaction for ^{27}Al and magnetic measurements[10,11] have shown that the easy magnetization axis lies along the [111] axis. This orientation of the magnetic moments is preserved at all temperatures below T_c.

The hyperfine interaction for Sn impurity atoms in the $GdAl_2$ lattice should have features dependent on the nature of the impurity localization. The magnetic dipole field (B_d) and the electric field gradient (EFG) are zero for a Gd site with cubic symmetry. Consequently, the hyperfine interaction for Sn impurity atoms on Gd sites will be determined only by the magnitude of the magnetic hyperfine field B_{hf} directed along the [111] axis. Both the EFG and B_d will differ from zero if the impurity atom is localized on Al sites. The structurally equivalent Al sites are essentially nonequivalent from the point of view of the parameters of the hyperfine interaction. This nonequivalence arises, firstly, because of different orientation of the axes of the EFG tensor relative to the direction of the magnetic hyperfine field and, secondly because of the difference in magnitude and orientation of the anisotropic contribution to MHFI. One of the reasons for an anisotropic contribution to the local magnetic field is the anisotropy of the dipolar magnetic field.

Two groups of nonequivalent Al sites (sites *a* and *b* in Fig. 1) with population ratio $a{:}b = 1{:}3$ arise if the magnetization is directed along the [111] axis in the $GdAl_2$ lattice. The principal axis of the axisymmetric EFG tensor for *a* sites (the threefold symmetry axis) coincides with the magnetization direction (and consequently with the B_{hf} direction). For *b* sites, the principal axis of the EFG tensor makes an angle $\theta = 70^\circ\,32'(\cos\theta = 1/3)$ with the B_{hf} direction. As will be seen below, the magnetic hyperfine interaction energy for Sn in $GdAl_2$ is much greater then the quadrupole interaction energy. In this case the quadrupole shift of the hyperfine structure component in the Mössbauer spectrum of ^{119}Sn is

$$\Delta E_Q = \pm (e^2qQ/4)\,(3\cos^2\theta - 1)/2, \qquad (1)$$

where Q is the nuclear quadrupole moment, q = EFG. The $+(-)$ sign corresponds to γ transitions between levels $m = \pm 3/2 (m = \pm 1/2)$. It follows from Eq. (1) that the quadrupole shifts for the two extreme components of the magnetic hyperfine structure (transitions between levels $m = +3/2$ and $m = -3/2$) are

$$\Delta E_Q(a) = e^2qQ/4 \quad (\text{sites } a),$$
$$\Delta E_Q(b) = -e^2qQ/12 \quad (\text{sites } b). \qquad (2)$$

The quadrupole shift for *b* sites is thus one-third that for sites *a* and has opposite sign.

The components of the anisotropic dipole field for Al on sites *a* and *b* in the $GdAl_2$ lattice have been calculated.[9] The theoretical value of the contribution B_d to MHFI for *a* sites was found to be -0.7 T. This contribution for *b* sites is positive and about three times smaller in absolute magnitude. The experimental values of the anisotropic contribution to MHFI for ^{27}Al were very close to the calculated values of B_d.[9,10] We should note that this agreement cannot occur in the general case since the anisotropic contribution to MHFI can be determined not only by the magnitude of B_d but also by the anisotropy of the polarization of the outer electrons of the atom.

As will become evident later, both the nature of the localization of an Sn impurity atom in the $GdAl_2$ lattice and the signs of the EFG and of the anisotropic contribution to MHFI could be determined from these features of the hyperfine interaction.

3. PREPARATION OF $GdAl_2$ ALLOYS, THE MEASUREMENT METHOD, AND THE MAIN RESULTS

The $GdAl_2$ alloy was prepared by melting the components in an arc furnace in an argon atmosphere. The melting was repeated several times to improve the homogeneity of the alloy. Aluminum of 99.999% purity and gadolinium of not worse than 99.9% purity was used. Metallic tin, enriched to 92% with the isotope ^{119}Sn was previously introduced into the aluminum. The tin impurity concentration in the alloy was 0.4 at. %.

The $GdAl_2$ alloy is formed in a well ordered state directly from the melt. NMR data[9] show that annealing of specimens after melting has no noticeable influence on the parameters of the hyperfine interaction for ^{27}Al. Nevertheless, we measured the Mössbauer absorption spectra in $GdAl_2$ powders prepared immediately after melting and after annealing the ingots at 750 °C for 50 h. We could find no noticeable difference in the parameters of spectra measured before and after annealing. The localization of Sn atoms in $GdAl_2$ is thus stable and is not changed by heat treatment of the alloy.

Mössbauer absorption spectra of 23.9 keV ^{119}Sn γ rays were measured in the temperature range from 6 to 300 K with $CaSnO_3$ sources, using spectrometers with constant acceleration. The temperature of the resonant absorber was maintained constant to an accuracy of 0.1 K. A resonant $CaSnO_3$ detector was used to register the Mössbauer radiation for temperatures below 77 K. The increased resolving power of such a detector was used to study some line-shape features caused by quadrupole interaction and by the anisotropic contribution to MHFI. The absolute calibration of the spectrometer was carried out using a laser interferometer.

A Mössbauer spectrum measured at 6 K is shown in Fig. 2. The magnetic hyperfine structure corresponding to

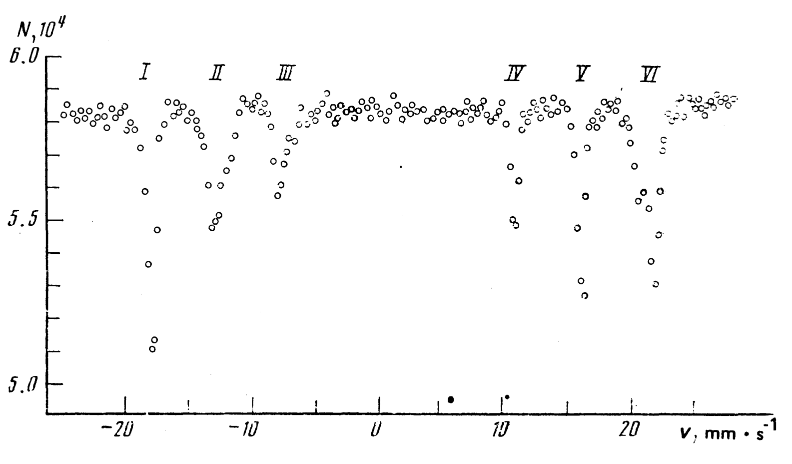

FIG. 2. Mössbauer spectrum for ^{119}Sn impurity atoms in $GdAl_2$, measured at 6 K.

the magnetic hyperfine field $B_{hf}(6\text{ K}) = -29.3 \pm 0.1$ T is well resolved. The sign of B_{hf} was determined from the shift in the components of the hyperfine structure in an external magnetic field 0.8 T at 77 K. The temperature dependence $B_{hf}(T)$ is shown in Fig. 3. The Curie temperature found from this dependence is 169 ± 2 K, which agrees with the known result for pure $GdAl_2$. This agreement can be regarded as an indication of the formation of a solid solution of tin in $GdAl_2$. A partially resolved quadrupole doublet was observed in the paramagnetic phase (in the temperature range from 172 to 300 K). The quadrupole splitting in this temperature range is constant; the magnitude of the splitting corresponds to an absolute magnitude of the quadrupole interaction constant

$$|e^2qQ| = 1.36 \pm 0.20 \text{ mm/s}. \tag{3}$$

The isomer shift is 1.90 ± 0.03 mm · s^{-1} at 6 K and 1.86 ± 0.02 mm · s^{-1} at 293 K (relative to $CaSnO_3$ at room temperature).

It is easy to see that the spectrum shown in Fig. 2 does not have with respect to its center the symmetry that should be observed for pure magnetic hyperfine interaction. This asymmetry, produced by the combined effect of quadrupole interaction and anisotropic magnetic interaction, will be discussed in the next section of the present work.

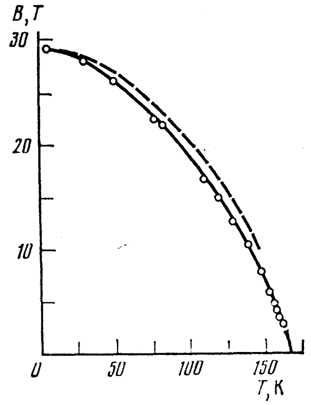

FIG. 3. Temperature dependence of magnetic hyperfine field $B(T)$ for ^{119}Sn impurity atoms in $GdAl_2$. The dashed line shows the temperature dependence of spontaneous magnetization[8] normalized to the value of the hyperfine field at 6 K.

In concluding this section, we note two more features of the spectra which will not be discussed later on:

1. A single broadened line, the area under which amounted to about 15% of the overall width of the spectrum, was observed in the spectra at temperatures above 30 K besides the magnetic hyperfine structure component. The presence of this line evidently means that the corresponding fraction of Sn impurity atoms do not form a solid solution and are contained in a contaminant phase. At temperatures below ~100 K this phase goes over to a magnetically ordered state which manifests itself in an increase in line width with subsequent splitting at low temperatures. The presence of the contaminant phase does not show up in the spectrum of Fig. 2 due to the small intensity of this line and its splitting. The presence of the contaminant phase was not an appreciable influence on the accuracy in determining the hyperfine interaction parameters, thanks to the well resolved hyperfine structure of the fundamental spectrum.

2. Some smearing of the phase transition from the ferromagnetic to the paramagnetic state was observed: the paramagnetic line in the spectrum appeared already at a temperature ~155 K, i.e., nearly 15 K below the nominal T_c. To explain this smearing it can be assumed that the Sn impurity atoms are a destructive influence on the exchange interaction in $GdAl_2$, so that the real T_c in regions of enhanced local concentration of tin is lower than the nominal value. This explanation is not, however, the only possiblity. Attention must be called to the results[12] of a study of the reasons for an anomalous T_c for $GdAl_2$ alloys. It was noted that T_c was close to 153 K for some alloys, appreciably below the "normal" value 170 K. The reduction in T_c was not due to a departure from stoichiometry or to the presence of impurities. The conclusion was drawn, on the basis of results of x-ray analysis, that the magnetic properties of $GdAl_2$ are sensitive to local structural disorders, in particular to the destruction of the ideal ordered position of the Gd and Al atoms in the corresponding lattice sites. It is possible that the smearing of the magnetic phase transition, which is observed in our measurements, is also a reflection of local imperfections in the crystal structure.

4. DISCUSSION OF THE RESULTS

4.1. Localization of Sn atoms, the electric field gradient and the anisotropic contribution to the local magnetic field

An unambiguous conclusion about the localization of Sn impurity atoms on Al sites can be drawn from a comparison of the measurements with the features of the hyperfine interactions in $GdAl_2$ considered above (Sec. 2). By assuming only such a form of localization, the features of the shape of the Mössbauer spectrum at low temperatures and the nonzero quadrupole interaction in the paramagnetic phase can be explained.

Unfortunately, a detailed quantitative analysis of the form of the components of the hyperfine structure is not possible, since the shifts of the lines produced by quadrupole interaction and by the anisotropic magnetic field are very small, and the structure of the components is not resolved. The shift of the components is especially small for *b* sites, which determine 75% of the overall intensity of the spectrum. It follows from a comparison of equality (3) with Eq. (2) that the quadrupole shift for Sn atoms on *b* sites is close to $0.1\ \mathrm{mm\cdot s^{-1}}$, which constitutes only 10% of the line width of the spectrum. It is, in addition, difficult to take account of a possible line broadening related to scatter of the parameters of the hyperfine and of the Lorentz field. Such a scatter can arise from structural imperfections, from the presence of impurity atoms, and also from variations in the dimensions and shape of the particles of the resonant absorber. This broadening is, naturally, small but for Sn atoms in *b* sites it can be comparable with the shift of the hyperfine structure component. In this case we confine ourselves to an approximate interpretation of the spectrum, considering only the shifts of the components for Sn atoms on *a* sites and completely neglecting these shifts for *b* sites. Such an approximation is sufficient to explain the main spectral features and allows us to obtain concrete information on the quadrupole interaction and the anisotropic contribution to the magnetic field. When approximating the spectrum by a sum of Lorentz lines, the free parameters in this approximation are the isotropic hyperfine field B_{hf}, the quadrupole and magnetic anisotropic shift of the components for *a* sites, and also the line width and isomer shift. Since aluminum *a* and *b* sites are structurally equivalent, one can assume that the isomer shift is the same for Sn atoms in these two sites.

It is not difficult to show that the observed asymmetry of the Mössbauer spectrum and the features of the shape of the components of the hyperfine structure are explained by the combined influence of quadrupole interaction and the anisotropic component of the local magnetic field for Sn atoms in aluminum *a* sites. This conclusion follows directly from a comparison of the parameters of lines situated (to a first approximation) symmetrically relative to the center of the spectrum. It can be seen that line I lies higher and is narrower than line VI (see Fig. 2). A similar relation between the parameters is observed for the pairs of lines II and IV, III and IV, but in this case the greater height and smaller width occur for the lines situated at the higher energy. We recall that the magnetic and quadrupole interactions do not have the same influence on the position of such symmetrically-placed lines on an energy scale. According to Eq. (1), the quadrupole shift for such lines has the same sign, while the change of local magnetic field produces their shift in opposite directions. It is thus clear that whereas these shifts are summed for one of the lines, they should be subtracted for the other. The small width of line I is then explained by almost exact compensation of the quadrupole shift and the shift due to the anisotropic magnetic interaction for *a* sites. As a result of this compensation, the position of the *a* component of line I practically coincides with the position of component *b*. The shifts are additive for line VI so that the existence of the *a* and *b* components shows up separately in the spectrum. The intensity of the component lying at the lower energy (about 25% of the total intensity of line VI) agrees well with what is expected for Sn atoms on *a* sites. The relations between the parameters for the pairs of lines II–V and III–IV are similarly explained, the only difference being that the sign of the quadrupole shift is opposite (see Eq. (1)) for these lines. Since the anisotropic contribution to the MHFI for *a* sites decreases the total splitting energy, the sign of the anisotropic component of the local magnetic field (B_A) must be positive (opposite to the sign of the magnetic hyperfine field B_{hf}). Cancellation of the magnetic and quadrupole shifts for line I indicates then that the quadrupole interaction constant e^2qQ is negative.

The following values of the parameters were obtained as a result of approximating the spectrum measured at 6 K by a sum of Lorentz lines:

$$B_{hf}=-29.3\pm0.1\ \mathrm{T},$$

$$e^2qQ=-1.4\pm0.4\ \mathrm{mm\cdot s^{-1}},B_A(a)=+0.7\pm0.1\ \mathrm{T}.$$

The uncertainties connected with the approximation in the model (neglect of the quadrupole shift and of the anisotropic component of the local magnetic field for *b* sites) are included in the errors for these quantities. It was assumed that the isotropic hyperfine field B_{hf} was the same for *a* and *b* sites.

The value of the quadrupole interaction found at 6 K agrees with its value at high temperatures (Eq. (3)), which is confirmed by the weak temperature dependence of EFG. If the value of this constant in Eq. (3) is used, then for $Q = -0.06$ barn[13] we find for EFG in the region of the Sn nucleus;

$$q=(+18\pm3)\,10^{17}\ \mathrm{V/cm^2}.$$

This value is several times larger than values of EFG usually found for Sn in noncubic metals.

As was mentioned in Sec. 2, the calculated value of dipole field for *a*Al sites in $GdAl_2$ is close to -0.7 T. The positive sign of B_A found by us indicates that for Sn impurity atoms the dominant contribution to the anisotropic magnetic interaction is not determined by the dipole field, but by anisotropic polarization of the electron shells of the atoms.

4.2. Magnetic hyperfine field and its temperature dependence

The magnetic hyperfine field for Sn impurity atoms in $GdAl_2$, $B_{hf} = -29.3$ T, turned out to be unexpectedly large, although slightly less than for Sn in pure gadolinium

($B_{hf} = -32.9$ T, Ref. 2). This result was unexpected because, in comparison with metallic gadolinium, the alloy $GdAl_2$ is much diluted in the magnetic sense. The magnetic moment of Gd atoms in pure gadolinium and in $GdAl_2$ are almost the same, but impurity atoms in a Gd matrix have 12 neighboring Gd atoms in the first coordination sphere, while for Al sites in the $GdAl_2$ matrix there are only 6 such neighbors. Both the mean atomic magnetic moment of the matrix and the magnetic moment of the first coordination sphere are thus appreciably less in $GdAl_2$ than in pure gadolinium.

In view of the extremely scanty experimental results, it would be premature to draw unambiguous conclusions about the reasons for such a sharp lack of correspondence between the hyperfine field and the magnetic moment in these two matrices. We suppose that it is an indication of a strong radial dependence of MHFI. We should note that in pure gadolinium the 12 nearest neighbors are located at a mean distance of 3.605 Å, while in the $GdAl_2$ matrix the distance between an Sn impurity atom and the six nearest Gd atoms is 3.271 Å. It is known that for an Sn atom in alloys of transition *d*-metals such a reduction in distance leads to a sharp increase in the negative contribution to the magnetic hyperfine field.[1] It is not impossible that a similar situation can occur in ferromagnetic alloys of rare earth elements.

It is quite probable that the radial dependence of MHFI manifests itself also in the temperature dependence of the hyperfine field. The results shown in Fig. 3 show that on raising the temperature, B_{hf} falls faster than the spontaneous magnetization in $GdAl_2$. A simple and natural explanation for such a temperature "anomaly" is based on a consideration of the dependence of hyperfine field on the thermal expansion of the crystal lattice (see, for example, Ref. 14). If an increase in interatomic distance as a result of thermal expansion is accompanied by a reduction in the negative contribution to B_{hf}, then on raising the temperature the negative hyperfine field must fall faster than the magnetization. Just such a sign for the temperature anomaly in B_{hf} is observed experimentally. One must have data on the radial dependence of contributions to B_{hf} for a quantitative evaluation of this effect, but such data for Sn in rare earth magnetic materials are so far unavailable. We propose to carry out similar experiments with other intermetallic materials having different radial distributions of the magnetic moments, to obtain more concrete information on the radial dependence of MHFI.

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