

Exchange and hyperfine interactions in the magnetic intermetallic compounds $R_2\text{In}$

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Mössbauer spectroscopy has been used to study the effect of intraionic fd exchange on the magnetic hyperfine interaction for ^{119}Sn impurity atoms in the magnetic intermetallides $R_2\text{In}$ ($R = \text{Nd, Sm, Gd-Tm}$). The anomalously high values of the magnetic hyperfine field and of the Curie temperature in the compounds Nd_2In and Sm_2In are interpreted as a consequence of a sharp increase in the degree of hybridization of the wave functions of the magnetic $4f$ electrons and the outer electrons. This effect is produced by the nonlinear dependence of the radius of the $4f$ electron distribution on the atomic number of the element R . The indirect RR exchange constant in the compound Nd_2In is almost twice as great as in the compounds with heavy R elements ($R = \text{Gd-Tm}$). The influence of the orbital moment of the R^{3+} ion in $R_2\text{In}$ compounds on the exchange and hyperfine interactions is negligibly small. The orientation of the magnetic moments of the R^{3+} ions in $R_2\text{In}$ compounds was determined. In the temperature range 30–80 K an orientational spin transition was observed for Dy_2In . The magnetic transition at the Curie temperature in the compound Nd_2In is a first order phase transition.

1. INTRODUCTION

The large variety of magnetic properties of intermetallic compounds and alloys of rare-earth elements is due to the competition between exchange interactions of different type. This competition is even evident in such systems in which the rare-earth (R) ions are the only source of exchange polarization of the electrons. Compounds with one and the same structure often have appreciably different magnetic properties, which can change both on changing the type of R ion and also on changing the temperature. In rare-earth magnetic materials direct overlap of the wave functions of the magnetic $4f$ electrons is absent (or is very small), so that the RR exchange interaction should have the character of indirect exchange. Until recently it was conventional to discuss RR exchange within the framework of a long-range (oscillatory) interaction via the conduction-band s electrons (RKKY interaction). Many results have been obtained recently which contradict this model, therefore the applicability of the RKKY theory for explaining the magnetic properties of intermetallides of the rare earths has come into question. An alternative point of view is based on the results of calculations of the band structure, according to which the conduction band in rare-earth metals has a predominantly d character and is in this respect analogous to the conduction band of transition metals of the iron group (see, for example, Freeman and Troper *et al.*¹ and the references given therein). The RR interaction can then come about via the inner $5d$ electrons, polarized as a result of intraionic fd exchange, the energy of which is appreciably greater than the energy of fs exchange. The degree of localization of $5d$ electrons is greater than of s electrons so that in the framework of such a concentration the RR exchange interaction should be of a more local character than follows from the model of RKKY interaction. Such parameters as the mean electron concentration or the magnitude of the s electron wave vector (which is a determining factor in the RKKY theory) lose their meaning. At the same time the features of the interatomic interaction at small distances and the details of the elec-

tron density distribution can take on the dominant role for atoms participating in indirect exchange.

The new concept of indirect RR exchange should be considered as a possible alternative which requires experimental confirmation. Results which can be obtained by traditional macroscopic methods are insufficient for a unique choice of a model of exchange interaction. A deeper understanding of the mechanisms for indirect RR exchange presupposes availability of information on the local character, which should directly reflect the interaction between exchange electron polarization and the features of the electronic structure of compounds. In this respect the most effective methods are based on a study of hyperfine interactions. The parameters of the hyperfine interaction for non-magnetic atoms in rare-earth magnetic materials are determined by the electronic and magnetic characteristics of the immediate surroundings,² so that the parameters are sensitive indicators of electron polarization produced by exchange interaction at small distances. Results recently obtained^{2,3} on hyperfine interactions for Sn impurity atoms in intermetallides of the rare earths confirm the high sensitivity of the hyperfine interaction parameters to the features of the electronic and magnetic structure of the compounds. Analysis of these results showed, in particular, that in rare earth magnetic materials the hyperfine interaction for a non-magnetic impurity atom has a clearly defined local character. The observed magnetic hyperfine fields are proportional to the polarization of the electrons on hybridized orbitals, formed on the interaction of the nonmagnetic atom with its nearest magnetic neighbors (R -ions). In this situation it is natural to expect a direct relation between the electron polarization, which determines the magnetic hyperfine interaction, and the characteristics of the indirect exchange interaction.

In the present work we have used the method of a Mössbauer nonmagnetic probe to study hyperfine interactions in magnetic intermetallic compounds $R_2\text{In}$ for ^{119}Sn impurity atoms (which replace In atoms in these compounds). $R_2\text{In}$ compounds with one and the same structure are formed with

almost all rare-earth elements. This provides a favorable situation for studying the influence of small variations in electronic structure, associated with the change in the type of R ion, on RR exchange and on the hyperfine interaction parameters. New results on the magnetic properties of $R_2\text{In}$ compounds, obtained by Mössbauer spectroscopy, are given in Sec. 3 below. The results of the joint analysis of the magnetic characteristics of $R_2\text{In}$ compounds and of the hyperfine interaction parameters are presented in Sec. 4. A model to explain the anomalous changes in the exchange interaction and magnetic hyperfine fields in $R_2\text{In}$ compounds is proposed, based on a consideration of the dependence of valence electron polarization on features of the spatial distribution of electron density.

2. PROPERTIES OF $R_2\text{In}$ COMPOUNDS

$R_2\text{In}$ compounds have the hexagonal N_2In structure.⁴⁻⁶ The lattice constant depends linearly on the atomic number of the R element and decreases by about 4% in the sequence $\text{Nd}_2\text{In}-\text{Tm}_2\text{In}$. The In atoms (and Si impurity atoms) lie at the centers of trigonal prisms and have 11 R^{3+} ions as nearest neighbors (situated at several different distances). All positions of the In atoms are structurally equivalent. The compounds have a relatively high magnetic-ordering temperature (up to 190 K for Gd_2In). Results of magnetic measurements^{5,6} showed that at low temperatures most of the compounds are ferromagnetic, but indications were obtained of various forms of magnetic ordering in different temperature ranges for some of them. The magnitudes of the effective magnetic moments in the paramagnetic phase are close to the theoretical values for free R^{3+} ions. On the whole, information on the magnetic properties of $R_2\text{In}$ compounds is very incomplete and in part contradictory. In particular, results on the magnetic spin structure are absent, the types of magnetic phase transitions have not been determined, and in a number of cases the type of magnetic ordering has not been established. The temperature dependences of the magnetization are often of anomalous form,^{5,6} but these anomalies are difficult to interpret on the basis of macroscopic measurements alone. The method of a Mössbauer nonmagnetic probe enables a measurement to be carried out without an external magnetic field, which renders the results independent of macroscopic magnetization of the specimen.

The intermetallics $R_2\text{In}$ ($R = \text{Nd}, \text{Sm}, \text{Ga}-\text{Tm}$) were obtained by arc melting in argon. In each melt, between 0.3 and 0.8% of the indium was replaced by tin enriched in the isotope ^{119}Sn . The formation of the Ni_2In type structure was confirmed by x-ray analysis. $R_2\text{In}$ intermetallics are formed in a well ordered state immediately after melting; annealing at a temperature of about 800 K had no appreciable influence on the parameters of the spectra. Mössbauer absorption spectra were measured in the temperature range from 5 K to a temperature exceeding the Curie point, and a CaSnO_3 radioactive source (which was at room temperature).

Good resolution of the magnetic hyperfine structure was observed in the spectra for all the compounds. The widths of the components of the hyperfine structure were equal to 0.9–1.4 mm/s, which points to the absence of appreciable scatter in the hyperfine interaction parameters and confirms the production of well ordered intermetallics. The standard procedure of approximating a spectrum by a sum

of Lorentz lines was applied to analyze the spectra. The quadrupole shift of the hyperfine structure components was very small so that it could be taken into account within the framework of perturbation theory. The sign of the magnetic hyperfine field B_{hf} for the compounds with $R = \text{Nd}, \text{Tb}, \text{Dy}, \text{Tm}$ was determined from the shift of the components of the hyperfine structure in an external magnetic field. (The sign of B_{hf} for Gd_2In was determined earlier.²) As was expected, B_{hf} is positive (i.e., parallel to the total magnetization) for compounds with light R elements ($J = L - S$ coupling scheme) and negative for compounds with heavy R elements ($J = L + S$ coupling scheme). This indicates that (as in other rare-earth intermetallics^{2,3}) B_{hf} always has a negative sign relative to the direction of the spin of the R^{3+} spin.

The magnetic hyperfine field and the quadrupole and isomer shifts, measured at 5 K, are shown in Table I. The quadrupole shift component of the hyperfine structure is determined as

$$\Delta = e^2qQ(3\cos^2\theta - 1)/8, \quad (1)$$

where q is the electric field gradient (EFG) in the region of the nucleus, Q is the nuclear quadrupole moment, and θ is the angle between the principal axis of the EFG tensor and the direction of the magnetic hyperfine field. For Sn atoms replacing In atoms in $R_2\text{In}$ compounds a small axially symmetrical EFG should be observed; the principal axis of the EFG tensor coincides with the hexagonal crystallographic axis. It follows from geometrical considerations that the EFG should have a positive sign. Since $Q < 0$ for ^{119}Sn , a negative hyperfine interaction constant ($e^2qQ < 0$) corresponds to a positive EFG. The negative sign of the shift Δ will then correspond to a preferential orientation of B_{hf} (and, consequently, of the magnetic moments of the R^{3+} ions) along the hexagonal axis; for a positive shift Δ the moments are arranged in the basal plane. As can be seen from Eq. (1), the quadrupole shift for $\theta = 0^\circ$ should be twice as large as for $\theta = 90^\circ$ in absolute magnitude.

3. MAGNETIC PHASE TRANSITIONS AND MAGNETIC STRUCTURE

3.1. Temperature dependences of B_{hf} and the nature of the magnetic phase transitions

The temperature dependences of the magnetic hyperfine field $B_{\text{hf}}(T)$ in the majority of compounds had no anomaly and were close to the corresponding Brillouin functions. The $B_{\text{hf}}(T)$ dependences for the compounds with $R = \text{Nd}, \text{Sm}, \text{Tb}, \text{Dy}$ and Tm are shown in Figs. 1 and 2 as examples. The Curie temperature T_C found from these dependences are shown in Table I. The values of T_C found agree satisfactorily on the whole with the results of magnetic measurements^{5,6} (which, incidentally, are tentative estimates in a number of cases).

The closeness of the $B_{\text{hf}}(T)$ dependences to the Brillouin functions in the whole temperature range indicates that the magnetic phase transitions are continuous phase transitions (second-order transitions). The compound Nd_2In is an exception, and its $B_{\text{hf}}(T)$ dependence is shown in Fig. 1. This dependence has two features: an anomalous decrease of B_{hf} in the range 25–40 K and a discontinuous disappearance of B_{hf} on raising the temperature to 111 K. The second of these features corresponds to the magnetiza-

TABLE I. Magnetic hyperfine fields B_{hf} , quadrupole shifts Δ , and isomer shifts for ^{119}Sn impurity atoms in intermetallic compounds R_2In . The Curie temperatures T_C are determined from the temperature dependence of the magnetic hyperfine fields.

R	B_{hf} , kOe (± 1 kOe)	Δ , mm/s (± 0.03 mm/s)	Isomer shift mm/s (± 0.03 mm/s)	T_C , K (± 1 K)
Nd	+219.0	-0.09	1.97	111
Sm	(+)273.3	+0.05	1.95	160
Gd	-373.4	+0.07	1.95	198
Tb	-328.2	+0.07	1.92	172
Dy	-271.2	+0.06	1.87	131
Ho	(-)222.6	-0.09	1.92	91.5
Er	(-)151.8	-	1.92	50
Tm	-84.1	+0.06	1.87	23.5

Note: The isomer shift, B_{hf} , and Δ are measured at 5 K. The isomer shift is determined relative to a room-temperature CaSnO_3 source. The sign of B_{hf} is shown in parentheses.

tion tending to zero discontinuously and indicates that the magnetic transition in Nd_2In is a first order phase transition. On lowering the temperature, a transition is observed at 110 K, i.e., it is characterized by a hysteresis width of about 1 K.

It is natural to attribute the anomalous reduction in B_{hf} at temperatures 25–40 K to a weak noncollinear magnetic structure arising in Nd_2In . The negative sign of the quadrupole shift Δ (see Table I) means that at low temperatures the magnetic moments of the Nd^{3+} ions are oriented along the hexagonal axis. The reduction in the hyperfine magnetic field in the range 25–40 K corresponds to a deviation of the moments from this axis by an angle of about 20° . The reasons for the anomalous magnetic susceptibility, found by Gammari-Seale *et al.*,⁵ can be explained by these results. The temperatures at which sharp changes in susceptibility are observed correspond closely to the region of formation of noncollinearity and to the first order magnetic transition at the Curie temperature.

3.2. Magnetic structure and orientation of the magnetic moments

As can be seen from Table I, the quadrupole shift for most of the compounds ($\text{R} = \text{Sm}, \text{Gd}, \text{Tb}, \text{Dy}, \text{Tm}$) is positive, which corresponds to an arrangement of the magnetic moments in the basal plane. For Tb_2In this result agrees with results obtained by the method of neutron diffraction.⁷ In the case of Nd_2In and Ho_2In the quadrupole shift is negative and, consequently, the magnetic moments are oriented

along the hexagonal axis (i.e., perpendicular to the basal plane). A spin orientational transition was observed for the compound Dy_2In in the temperature range 30–80 K. As can be seen from Fig. 2, the quadrupole shift at low temperatures is positive, which corresponds to location of the moments in the basal plane. As the temperature is increased, a gradual rotation of the moments is observed, and at temperatures above 80 K the magnetic moments are oriented along the hexagonal axis. In agreement with Eq. (1), the quadrupole shift at temperatures above 80 K is about twice as large in absolute magnitude as at low temperatures.

The values of B_{hf} in compounds R_2In (see Table I) are typical for Sn impurity atoms in compounds of the rare earths with ferromagnetic orientation of the magnetic moments of the R^{3+} ions.^{2,3} The ferromagnetic configuration of the moments in the immediate neighborhood of a Sn atom is confirmed both by the high values of the ratio B_{hf}/S_z (S_z is the projection of the spin of the R^{3+} ion) and by the constancy of this ratio in the series of compounds Gd_2In – Er_2In . At the same time, measurements of the magnetization in weak fields showed^{5–7} that at low temperatures transitions are observed in Gd_2In and Sm_2In to a state with weak magnetization, characteristic of antiferromagnetic ordering. We did not find any anomalies in the temperature dependences $B_{\text{hf}}(T)$ for these compounds at temperatures corresponding to such transitions. It was suggested⁷ that for Gd_2In at low temperatures this compound goes over to a state with a spiral (helical) magnetic structure. Our results can be reconciled with this hypothesis if it is assumed that the period of

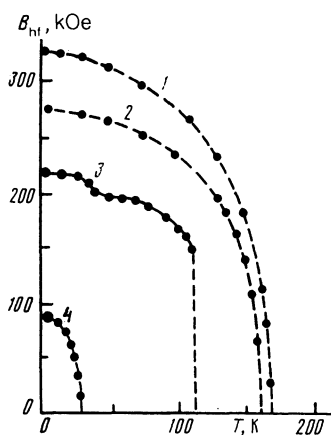


FIG. 1. Temperature dependences of the magnetic hyperfine field B_{hf} for ^{119}Sn impurity atoms in the compounds (1) Tb_2In , (2) Sm_2In , (3) Nd_2In , (4) Tm_2In .

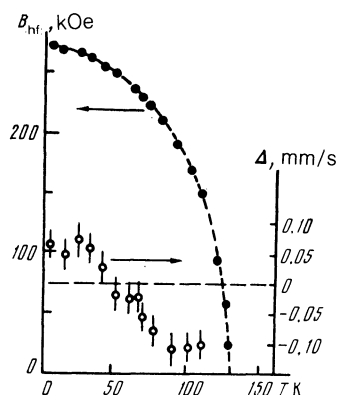


FIG. 2. Temperature dependences of the magnetic hyperfine field B_{hf} (●) and the quadrupole shift Δ (○) for ^{119}Sn impurity atoms in the compound Dy_2In .

the helix is very great. In that case the angle of rotation of the magnetic moments for neighboring elementary cells is very small and the configuration of the moments in the immediate vicinity of each of the Sn atoms will practically not differ from ferromagnetic. Since the hyperfine interaction for Sn impurity atoms in intermetallics of rare earths is local,^{2,3} the magnitude of B_{hf} is insensitive to a transition from a ferromagnetic structure to a spiral type of structure.

4. INFLUENCE OF A RADIAL DISTRIBUTION OF MAGNETIC 4f ELECTRONS ON EXCHANGE AND HYPERFINE INTERACTION

It is usual to assume that for a nonmagnetic atom in compounds and alloys of rare earths the magnetic hyperfine field is, to a first approximation, proportional to the projection of the spin of the rare earth ion S_z . A proportionality between B_{hf} and S_z for a number of isostructural compounds should arise if, firstly, spin exchange with the magnetic 4f electrons is the only source of electron polarization and, secondly, if the electronic structure of the compounds does not change with a change in the type of R ion. It is, however, known that strict proportionality between B_{hf} and S_z is not observed. The departures from proportionality are usually not very large; the reasons for these departures in the general case are not known. One of the probable reasons for such departures is the orbital contribution to electron polarization.

Results were recently obtained on the magnetic hyperfine interaction for Sn impurity atoms in ferromagnetic isostructural RAl_2 compounds.³ It was shown that the departures from proportionality between B_{hf} and S_z in this case can be explained by orbital polarization. In compounds RAl_2 the spin and orbital contributions to B_{hf} are of opposite sign, with the relative magnitude of the orbital contribution close to 20%. For compounds with heavy R elements the orbital is subtracted from the spin contribution, for compounds with light R elements the two contributions are summed. In this way, the departures from proportionality between B_{hf} and S_z in RAl_2 compounds are due to the destruction of the first of the two conditions for proportionality stated above.

As will be seen below, a quite different situation is realized for R_2In compounds in which the role of the orbital moment is negligibly small and the main factor determining the large variation in the ratios B_{hf}/S_z is the change in the radial distribution of the magnetic 4f electrons. The magnitudes of B_{hf}/S_z for Sn atoms in R_2In compounds are shown in Table II. It can be seen that for compounds with heavy R

elements (from Gd_2In to Er_2In) the ratio B_{hf}/S_z is constant to high accuracy. (The departure from the mean value of 106 kOe/spin does not exceed 5%.) At the same time, for compounds with light R elements this ratio is anomalously high and for Nd_2In it is almost twice as great. These behaviors cannot, apparently, be explained by the orbital contribution to B_{hf} . The largest change in the projection of the orbital moment L_z takes place for heavy R ions (from $L_z = 0$ for Gd^{3+} to $L_z = 6$ for Ho^{3+}), but it is just for compounds with these elements that B_{hf}/S_z is almost strictly constant. It must, therefore, be assumed that in R_2In compounds the electron polarization is completely (or almost completely) determined by spin exchange and the largest variations in B_{hf}/S_z are associated with changes in electron structure which lead to a sharp increase in electron polarization in compounds with $\text{R} = \text{Nd}, \text{Sm}$.

To verify this assumption, one must compare the magnitudes of B_{hf}/S_z with other properties of the compounds, which should also be determined by the degree of polarization of the valence electrons. Such a property is the indirect exchange interaction energy that determines the Curie temperature. If the energy of exchange interaction is determined solely by spin exchange with 4f electrons, then (in accord with the general theory of indirect exchange interaction and independent of the actual model of spin exchange) the Curie temperature should be proportional to the de Gennes factor G :

$$T_c \propto G(J) = (g-1)^2 J(J+1), \quad (2)$$

where J and g are respectively the total angular momentum and the g factor of the R^{3+} ion. In the sequence of compounds with unchanging electronic structure (and, consequently, with the same exchange interaction constant) the reduced Curie temperature $t_c = T_c/G$ should be constant; the variation of t_c should reflect changes in the exchange interaction energy, which depends on the type of R ion. Values of t_c are shown in Table II. The result of comparing the dependences of the values of B_{hf}/S_z and t_c on the atomic number of the R element is shown in Fig. 3. It can be seen that the two dependences are completely similar. This similarity confirms that the anomalous increase of B_{hf}/S_z and t_c in the compounds Nd_2In and Sm_2In is of one and the same nature and is due to a sharp increase in exchange polarization of the valence electrons. Taking into account the local character of B_{hf} , it is natural to suggest that the anomalous increase in t_c in compounds with light R elements is also due to a local mechanism dependent on features of the exchange interaction at small distances.

TABLE II. Parameters characterizing the hyperfine and exchange interaction in intermetallic R_2In compounds: the magnetic hyperfine field is normalized to the spin projection S_z of the R^{3+} ion; the reduced Curie temperature $t_c = T_c/G$ (G is the de Gennes factor; the root mean square radius of the 4f electron distribution r_{4f} (Ref. 8); the effective radius of the Wigner-Seitz cell r_N ; $\Delta r = r_N - r_{4f}$).

R	B_{hf}/S_z , kOe	T_c/G , K	r_{4f} , Å	r_N , Å	Δr , Å
Nd	178	60.3	0.528	1.763	1.235
Sm	154	35.9	0.496	1.747	1.251
Gd	107	12.6	0.468	1.735	1.267
Tb	109	16.4	0.458	1.720	1.262
Dy	108	18.9	0.450	1.714	1.264
Ho	111	20.3	0.440	1.707	1.267
Er	101	19.6	0.431	1.697	1.266
Tm	84	19.4	0.421	1.692	1.271

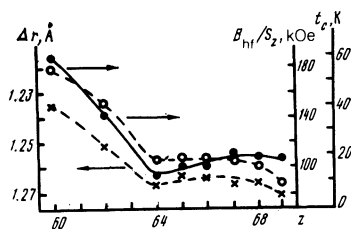


FIG. 3. Dependences, on the atomic number Z of the R element, of the values of (○) B_{hf}/S_z , (●) $t_c = T_c/G$ and (x) $\Delta r = r_N - r_{4f}$ in R_2In compounds.

Obviously, the observed changes of the exchange-interaction constant are not connected directly with the radial dependence of the indirect RR exchange, since the dependence of the interionic distance on the atomic number is linear and has no such singularities on going from light R-elements to heavy ones. In accord with the results of the theoretical calculations,¹ it is necessary to choose the factor that determines the indirect-exchange energy of the $5d$ electrons of the R^{3+} ion, the value of which is directly connected with the singularities of the interionic fd exchange.

The critical parameter of the model of intraionic exchange is the degree of overlap of the wave functions of the magnetic $4f$ and $5d$ electrons. There are no theoretical calculations of the dependence of such an overlap on atomic number, but at the qualitative level the problem can be considered in a semiempirical way. For this purpose parameters are used which are average characteristics of the electronic radial distribution. Such a parameter for $4f$ electrons is the root mean square radius $r_{4f} = \langle r_{4f} \rangle^{1/2}$. We will use below values of r_{4f} calculated by Freeman *et al.*⁸; these values of r_{4f} are given in Table II.

Any quantity which provides information about the ionic radius can be used to characterize the radial distribution of the outer $5d$ electrons. For analysis of the actual situation in R_2In compounds, we used the radius r_N which was defined as half the mean interatomic distance for R^{3+} ions (which is, in fact, equivalent to the radius of the Wigner-Seitz cell). The degree of overlap of the wave functions of the $4f$ and $5d$ electrons depends on the relation between the radii r_{4f} and r_N . For simplicity (bearing in mind the approximate nature of the analysis), the difference in the radii can be used as a parameter characterizing the degree of overlap

$$\Delta r = r_N - r_{4f}. \quad (3)$$

(The degree of overlap is larger the smaller the difference Δr .) The radii r_N and r_{4f} have an appreciably different dependence on the atomic Z . The magnetic $4f$ electrons in their mean position are considerably nearer to the nucleus than the outer $5d$ electrons, so that the radius r_{4f} depends more strongly on Z than does the radius r_N . As can be seen from the data of Table II, the change in r_N in the sequence of compounds R_2In is close to 4%, while the radius r_{4f} changes by about 25%. An even more important fact is that the $r_N(Z)$ dependence is linear, while the $r_{4f}(Z)$ dependence is appreciably nonlinear: when Z of light R elements is increased the radius r_{4f} decreases considerably faster than in the range of heavy elements.

The decisive role of this relationship becomes clear on comparing the $\Delta r(Z)$ dependence with the B_{hf}/S_z and t_c dependences already considered above (see Fig. 3). It can be

seen that all three dependences are similar while the $\Delta r(Z)$ relation reproduces remarkably well all the main features of the dependence of B_{hf}/S_z on the atomic number. This result should be regarded as confirmation of the model according to which the anomalous growth of B_{hf}/S_z and t_c for compounds with light R elements is determined by details of the intraionic fd exchange. The growth in the exchange polarization of the $5d$ electrons is a consequence of the increase in the radius r_{4f} relative to r_N , which leads to a more effective overlap of the electronic wave functions. This effect can be interpreted as a change in the degree of hybridization of the wave functions of the $4f$ and $5d$ electrons. In this connection one should take notice of the work of Fuggle *et al.*⁹ in which some alloys of the light R elements were studied by the method of photoelectron spectroscopy. The results of that work can be interpreted as an indication of the appreciable role of $4f$ - $5d$ hybridization, the degree of which falls with increasing atomic number.

Within the framework of the model of intraionic fd exchange the increase in B_{hf} and the raising of T_c for compounds with light R elements have one and the same cause and should be considered as a consequence of an increase in the degree of polarization of the $5d$ electrons of the R^{3+} ions. The degree of polarization of the $5d$ electrons determines both the indirect RR exchange constant (i.e., the Curie temperature) and also the magnitude of B_{hf} (through the polarization of the electrons on hybridized orbitals). The correctness of this conclusion becomes especially clear on comparing directly the values of B_{hf} and T_c within the framework of the general conception of indirect (spin) exchange. One can obtain between B_{hf} and T_c a relation in which the general dependence of these quantities on the degree of polarization of the electrons is eliminated. Recognizing that B_{hf} is proportional to the spin projection $S_z = J(g-1)$ while T_c is proportional to the factor $G(J)$ [Eq. (2)], one should expect the linear relation

$$B_{hf} [(J+1)/J]^{1/2} = K T_c^{1/2}, \quad (4)$$

where K is the proportionality constant. As can be seen from Fig. 4, Eq. (4) agrees well with the experimental results. In this representation, compounds with light R elements are in no way singled out, which is evidence of a single mechanism for indirect exchange over the whole sequence of R elements and confirms that in R_2In compounds exchange interaction really has the character of spin exchange.

We note that for Sn atoms in RA_2 intermetallides, representation of the experimental results³ in the form of Eq. (4) leads to quite a different result. In that case the linear

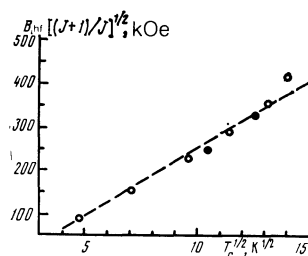


FIG. 4. Linear correlation between $B_{hf} [(J+1)/J]^{1/2}$ and $T_c^{1/2}$ (Eq. 4) in R_2In compounds. The full circles denote results for the compounds Nd_2In and Sm_2In with light R elements.

relation of Eq. (4) splits into two branches (light and heavy R elements), which reflects the existence of an orbital contribution to B_{hf} in RAl_2 intermetallides. At the same time, our results for cubic R_3In intermetallides are quite analogous to the results for R_2In compounds. An explanation of the reasons for the similarity and difference between the properties of the hyperfine interaction in the sequence of compounds of a different type is an interesting and timely problem. It should be expected that the application of the Mössbauer nonmagnetic probe to a wider circle of ferromagnetic and antiferromagnetic systems would enable new results to be obtained, essential for constructing a fuller picture of indirect exchange in rare earth magnetic materials.

¹A. J. Freeman, *Physica (Utrecht)* **B91**, 103 (1977), **B102**, 3 (1980), **B130**, 1 (1985); A. Troper, O. L. T. de Menezes, and A. Gomes, *J. Phys.* **F9**, 2457 (1979).

²N. N. Delyagin, G. T. Mujiri, V. I. Nesterov, and S. I. Reiman, *Zh. Eksp. Teor. Fiz.* **86**, 1016 (1984) [*Sov. Phys. JETP* **59** (3), 592 (1984)]; N. N. Delyagin, V. I. Krylov, V. I. Mujiri, and V. I. Nesterov, *Phys. Status Solidi* **B131**, 555 (1985).
³N. N. Delyagin, V. I. Krylov, N. I. Moreva, G. T. Mujiri, V. I. Nesterov, and S. I. Reiman, *Zh. Eksp. Teor. Fiz.* **88**, 300 (1985) [*Sov. Phys. JETP* **61** (1), 176 (1985)].
⁴A. Palenzona, *J. Less-Common Met.* **16**, 379 (1968).
⁵H. Gamari-Seale, T. Anagnostopoulos, and J. K. Yakinthos, *J. Appl. Phys.* **50**, 434 (1979).
⁶W. Bazela and A. Szytula, *J. Less-Common Met.* **138** (1), 123 (1988).
⁷S. P. McAlister, *J. Phys.* **F14**, 2167 (1984).
⁸A. J. Freeman and R. E. Watson, *Phys. Rev.* **127**, 2058 (1962); A. J. Freeman and J. P. Desclaux, *J. Magn. Magn. Mater.* **12**, 11 (1979).
⁹J. C. Fuggle, O. Gunnarsson, G. A. Sawatzky, and K. Schönhammer, *Phys. Rev.* **B37**, 1103 (1988).

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