Hyperfine fields at Fe^{57} nuclei in $Gd_xY_{1-x}Fe_2$ alloys

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The position of the center of gravity of the Fe^{57} NMR in $Gd_xY_{1-x}Fe_2$ has a discontinutiy as a function of the Gd concentration near the compensation point. This discontinuity can be attributed to a change in the magnetization of the conduction electrons. Analysis of the conduction-electron spin density distribution indicates that indirect exchange between the iron and gadolinium atoms takes place via delocalized *d* electrons.

Intermetallic compounds of rare earths with 3d transition elements are very promising magnetic materials from the point of view of practical applications. It is also important to investigate these substances in connection with the development of modern theoretical ideas concerning magnetism. The theory of indirect exchange via conduction electrons developed by Ruderman, Kittel, Kasua, and Yosida (the RKKY theory) is frequently invoked to explain the magnetic properties of such compounds[1]. However, this theory gives only qualitative agreement with experiment, owing, apparently, to the crudeness of the free-electron model.

The spin-density distribution of the collectivized electrons must be investigated in order to determine the type and characteristics of the interaction of the atomic spins with one another. We have investigated the nuclear magnetic resonance (NMR) of iron nuclei in $Gd_xY_{1-x}Fe_2$ compounds. The alloy specimens were prepared in the laboratory of O. P. Elyutin at the TsNIIChERMET (Central Scientific Research Institute for Ferrous Metallurgy)[2]. The NMR spectra were recorded at 4.2°K with a spin echo spectrometer. The first and second pulses were 10 μsec long; the pulses were applied at a repetition rate of 1 sec^{-1} with a delay of 60 μsec between the two pulses of a pair. Measurements were repeated on different batches of alloy over the entire range of composition.

The NMR spectra of the $GdFe_2$ and YFe_2 specimens each consists of two lines with a 3 : 1 intensity ratio, corresponding to the two inequivalent positions of the iron atoms in the crystal lattice. The hyperfine field strengths at the Fe^{57} nuclei were 240 ± 5 kOe for Fe I and 255 ± 5 kOe for Fe II in $GdFe_2$, and 220 ± 5 and 210 ± 5 kOe in YFe_2, in good agreement with previously published data[3]. The lines of specimens of intermediate composition broaden somewhat on approaching the composition x = 0.5, the broader lines being observed for the specimens with a low degree of Gd substitution; this indicates that there is a dipole contribution from the gadolinium atoms to the local field at the iron nuclei. Knowing the lattice constant and the structural type of the crystal, one can estimate the dipole contributions[4]. The calculations show that the dipole contribution from the gadolinium ions does not exceed 10 kOe. The fact that the line width reaches 2 MHz in the $Gd_{0.5}Y_{0.5}Fe_2$ specimen indicates that there is a contribution to the hyperfine field from the conduction electrons.

The manner in which the center of gravity of the NMR lines shifts with varying Gd content is shown in the figure. The figure also shows the gadolinium-content dependence of the magnetic moment of the iron as calculated from magnetization measurements on the assumption that the magnetic moment of Gd is 7.0 μ_B. It is evident from the figure that the hyperfine field varies in proportion to the magnetic moment except for a small but sudden step (discontinuity) near the gadolinium concentration $x_c = 0.43$. Our magnetic measurements showed that at 4.2°K the magnetic moment m_f of the gadolinium sublattice is equal to the magnetic moment m_d of the iron sublattice at this concentration, so that the saturation magnetization passes through a minimum as a function of the gadolinium concentration at $x_c = 0.43$. We find that $m_f > m_d$ when $x > 0.43$ and $m_f < m_d$ when $x < 0.43$. Thus, the discontinuity in the hyperfine field indicated by the figure takes place when the concentration passes through the magnetic compensation point x_c. This discontinuity in the hyperfine field strength can be related to a change in the magnetization of the conduction electrons near x_c. To see this let us write the following expression for the exchange energy of the two sublattices by analogy with the s-d exchange theory[5]:

$$\varepsilon(m_f, m_d, m_s) = -\tfrac{1}{2}A_{ff}m_f^2 - \tfrac{1}{2}A_{dd}m_d^2 - A_{fd}m_dm_f - A_{sf}m_fm_s - A_{sd}m_dm_s + \tfrac{1}{2}A_F m_s^2, \quad (1)$$

where m_f and m_d are the magnetizations of the f and d sublattices, m_s is the magnetization of the conduction electrons, the A_{ij} are the effective exchange parameters, and $A_F = N_s\mu_B/\chi_{Pauli}$. By minimizing this expression for the equilibrium values of the magnetization we can find an expression for the effective field acting on each of the sublattices:

$$H_{eff}^f = \left[A_{fd} + \frac{A_{sf}A_{sd}}{A_F} - A_{ff} - \frac{A_{sf}^2}{A_F}\right]m_f + \left[A_{ff} + \frac{A_{sf}^2}{A_F}\right]\varphi(x-x_c),$$
$$H_{eff}^d = [A_{dd} + A_{sf}^2/A_F - \xi A_{fd} - \xi A_{sf}A_{sd}/A_F]m_d + [A_{fd} + A_{sd}^2/A_F]\varphi(x-x_c), \quad (2)$$

where $\xi = (1 + A_{sd}/A_F)/(1 + A_{sf}/A_F)$ and $\varphi(x - x_c)$ is a function of the concentration that is odd about the point $x = x_c$.

Making use of the relation

$$m_s = \frac{A_{sf}m_f + A_{sd}m_d}{A_F} = \frac{A_{sf}}{A_F}\left[m_f + \frac{A_{sd}}{A_{sf}}m_d\right], \quad (3)$$

and assuming that $A_{sd} \sim A_{sf}$, we can obtain an equation of the form

$$m_s = R + K\varphi(x-x_c), \quad (4)$$

in which R and K are certain combinations of the effective exchange parameters. Equation (4) shows that the direction of the magnetization of the conduction electrons must change sign when the concentration passes

through the compensation point. Since the s-like conduction electrons have a finite density of states in the vicinity of the Fe nuclei, passage through the compensation point must clearly be accompanied by a sharp change in the hyperfine field. The observed change, however, is very small; this means that the density of states of collectivized s electrons at the iron nuclei is very low. Moreover, it is known[6,7] that the iron sublattice induces a considerable hyperfine field at the gadolinium nuclei, this induced field being in the positive direction (with respect to the direction of the Gd magnetic moment). At the same time, our measurements reveal no similar sudden change with concentration in the hyperfine fields at the Gd and Y nuclei[2].

Thus, we find that the collectivized electrons have a finite density at the gadolinium nuclei and a low density at the iron nuclei, i.e., they are described by p- or d-type wave functions that have nodes where the iron nuclei lie. This means that the exchange between the gadolinium and iron sublattices in $GdFe_2$ takes place via delocalized d electrons; this is in accordance with the conclusions reached in[8] for compounds of iron with silicon and aluminum. The hypothesis that the exchange between the d and f sublattices takes place via delocalized d electrons is also confirmed by the high Curie points of compounds of iron with rare-earth metals (780°K for $GdFe_2$). The Curie points of pure rare-earth metals, in which the exchange interaction takes place via the polarization of the conduction electrons, do not exceed 300°K.

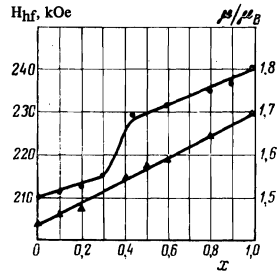
It is very interesting to compare the hyperfine fields at the iron nuclei in $Gd_xY_{1-x}Fe_2$ alloys and in Fe-Si and Fe-Al alloys[9,10]. In the table we give estimates of various contributions to the hyperfine field H_{hf} at the Fe^{57} nuclei under the condition that the proportionality constant between the hyperfine field H_{hf} and the magnetic moment μ_{Fe} is the same for the Fe-Si alloys as for the $Gd_xY_{1-x}Fe_2$ alloys. Here it is assumed that the hyperfine field at the iron nuclei can be expressed in the form

$$H_{hf}^{Fe}=H_{ep}+H_{neigh}^{Fe}+H_{neigh}^{Gd}+H_{intr}^{4s},$$

where H_{ep} is the hyperfine field due to exchange polarization of the s shell of the iron atom under the action of its magnetic moment; H_{intr}^{4s} is the hyperfine field associated with the polarization of the 4s electrons by the intrinsic magnetic moment of the atom; and H_{neigh}^{Fe} and H_{neigh}^{Gd} are the field induced by the neighboring iron and gadolinium atoms, respectively.

The agreement shown in the table between the calculated value of the hyperfine field at the Fe^{57} nuclei and its experimental value also confirms the assumption of exchange ordering of the magnetic moments via delocalized d electrons.

We note in this connection that the calculation of lattice sums by the RKKY interaction method leads to very low values for the Fermi energy ($E_f < 3$ eV). Calculations based on Friedel's theory[11] with the use of an effective mass of the d carriers would therefore be of interest.



Hyperfine field at the Fe^{57} nuclei (circles) and the magnetic moment of the Fe atoms (triangles) in $Gd_xY_{1-x}Fe_2$ versus Gd concentration x.

Contribution to the hyperfine field	According to [8]	Our results
Exchange polarization	−400 kOe	−310 kOe
Polarization of the 4s electrons by the intrinsic moment of the atom	+205 kOe	+160 kOe
Polarization due to neighboring iron atoms (via delocalized electrons)		
Contribution from the gadolinium ions	−145 kOe —	−110 kOe +20 kOe
Total:	−340 kOe	−240 kOe

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[1] K. N. R. Taylor, Adv. Phys. 87, 551 (1971).
[2] V. A. Vasil'kovskiĭ, N. M. Kovtun, A. K. Kupriyanov, S. A. Nikitin, and V. F. Ostrovskiĭ, Zh. Eksp. Teor. Fiz. 65, 693 (1973) [Sov. Phys.-JETP 38, 342 (1974)].
[3] G. J. Bowden, D. Bunbury, A. Guimaraes, and R. Snyder, J. Phys. 1C, 1376 (1968).
[4] G. Schulze, Metal Physics (Russ. Transl. Mir, 1971).
[5] S. V. Vonsovskiĭ, Magnetizm (Magnetism), Nauka, 1971.
[6] R. Gegenwarth, J. Budnick, S. Skalski, and J. Wernick, Phys. Rev. Lett. 18, 9 (1967).
[7] A. J. Freeman and R. Watson, in: Sverkhtonkie vzaimodeistviya v tverdykh telakh (Collected translated articles on Hyperfine Interactions in Solids), Mir, 1970, p. 99.
[8] Mary Beth Stearns, Phys. Rev. 4B, 4081 (1971).
[9] M. B. Stearns, Phys. Rev. 129, 1136 (1963).
[10] M. Rubinstein, G. H. Stauss, and M. B. Stearns, J. Appl. Phys. 31, 1334 (1966).
[11] J. Friedel, G. A. Leman, and S. Olszewski, J. Appl. Phys., 32, 325S, (1961).

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