MEASUREMENTS OF THE CONDUCTION-ELECTRON SPIN-DENSITY OSCILLATIONS IN FERROMAGENTIC ALLOYS

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The internal fields at various Fe^{57} sites in several ferromagnetic alloy series have been measured by Mössbauer-type experiments. These fields are proportional to the electronic spin density at an Fe^{57} nucleus. This spin density at the nucleus is composed of two contributions: one from the core s electrons, the other from the conduction s electrons. If we can choose an alloy system where the core contribution remains constant with variations of composition, then any changes in the internal field will be due to the changes in the conduction-electron spin density. Thus by observing the behavior of the internal field as a function of solute concentration of such a system, we can directly measure the radial dependence of the conduction-electron spin density. In the literature the most widely used forms for this variation are those derived by Ruderman and Kittel, and Yosida.[1] These measurements give a spin-density variation which has a magnitude seven times that predicted by the R-K form. Furthermore, the results seem to indicate that the conduction-electron spin susceptibility has a maximum for a wave vector near the Fermi-surface diameter as predicted by Overhauser.[2]

Previously we have measured the change in spin density for a nearest-neighbor ($N1$) model for Si and Al atoms in Fe.[3] These measurements indicate that in both these systems the core contribution does indeed remain constant over wide variations of solute concentration. We have now measured a number of other alloy series and have extended the computer program to include effects out to sixth-nearest neighbors (64 surrounding atoms). All the alloys used were body-centered cubic and were randomized to as great a degree as possible by the appropriate heat treatment. The analysis of the observed Mössbauer spectra is the following: With the aid of a computer we unfold the two outer peaks into all their components. For an $N6$ model there are $9\times7\times13\times25\times9\times7$ possible occupational configurations. The probabilities for these configurations are computed assuming complete randomness of the solute atoms. Each statistically significant component is then assigned a shift and shape. We varied the shape and found that a pure Fe shape was best. Consequently, shapes corresponding to pure Fe absorbers of the correct thickness were adopted. The field shift ΔH (relative to pure Fe) at a given Fe^{57} site arising from a specific occupational configuration of its neighborhood is assumed to be

$$\Delta H = \sum_{n=1}^{6} m_n \Delta H_n, \qquad (1)$$

where m_n is the number of nth nearest neighbors occupied by solute atoms, and ΔH_n is the contribution to the shift of a single solute atom at an nth-nearest-neighbor position. The computer program then determines the six values ΔH_1, $\Delta H_2, \cdots, \Delta H_6$ which minimize the mean-square deviation g^2 of the observed spectrum with respect to the calculated spectrum which is obtained by summing over all the components at the positions given by Eq. (1). The additivity assumption implied by Eq. (1) had been strongly evident in the works of reference 3. It was also checked here explicitly by separate $N2$- and $N3$-type analyses which allowed different shifts for each distinct configuration of first- and second-nearest neighbors. Additivity was again confirmed to a remarkable degree. Figure 1 shows a comparison of some typical experimental and calculated curves.

Table I gives the results for the FeAl series. For this series the values obtained are seen to be independent of concentration. From the behavior of the mean-square deviation we found that ΔH_1 through ΔH_4 are readily and accurately obtained. ΔH_5, while not as reliably obtained as the first four shifts, is reasonably accurate and well represented by the rms error shown. ΔH_6, being near a node and having only six atoms in its shell, is least accurate. It may also have a systematic error since it includes the average effect of all the more distant atoms. We find that to get reliable values for ΔH_1 and ΔH_2 at least four neighbors should be considered; also, in some systems ΔH_2 is quite concentration dependent. We therefore disagree with the results of Wertheim et al.[4] who found that ΔH_1 and ΔH_2 were adequate for analysis.

FeAl.—In Fig. 2 are shown the relative internal field shifts at an Fe atom as a function of the distance from an Al impurity atom. The right-hand

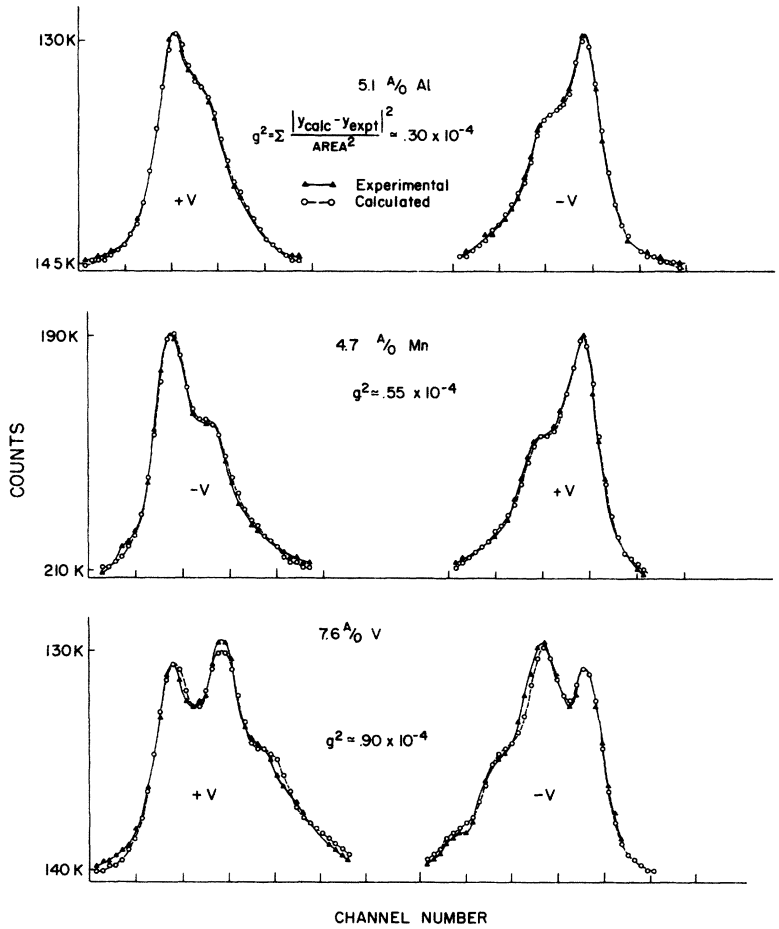

FIG. 1. Typical experimental and calculated curves of the outer peaks of the Mössbauer spectra. The experimental points have been omitted when they have the same value as the calculated points.

scale gives the corresponding indirect interaction energy in units of Mc/sec. Let us note some of the obvious features of the behavior of the internal field shifts of the FeAl series. For this alloy series the internal field shifts are independent of the Al content and the oscillations in the spin density seem to be about the pure Fe value. One would expect some variation of these quantities in most cases since the band structure and Fermi surface might vary somewhat with concentration. These results thus indicate that the Al atom is probably not contributing to the magnetic or band structure and acts very much like a hole in the Fe lattice. This confirms earlier conclusions on the negligible effect of Al on these properties in Fe alloys from heat-capacity measurements by Pessall et al.[5] From the position of the nodes in the spin-density curve, we find that the value

Table I. Percentage internal field shift for each type nighbor in the FeAl series.

Atomic % Al	$\Delta H_1/H_{Fe}$	$\Delta H_2/H_{Fe}$	$\Delta H_3/H_{Fe}$	$\Delta H_4/H_{Fe}$	$\Delta H_5/H_{Fe}$	$\Delta H_6/H_{Fe}$
5.1 Sample No. 1	−7.0	−4.0	+1.5	−0.3	−1.0	+0.3
Sample No. 2	−7.0	−4.1	+1.8	−0.4	−0.2	+0.2
10.6 Sample No. 1	−6.6	−3.4	+1.2	−0.4	−0.7	+0.5
Sample No. 2	−6.9	−3.4	+1.3	−0.2	−0.8	+0.2
13.2	−7.0	−3.7	+1.5	−0.3	−0.5	+0.4
15.2	−7.1	−3.8	+1.4	−0.2	−0.6	+0.2
Av. ±rms error	−6.9 ± 0.2	−3.7 ± 0.3	+1.4 ± 0.1	−0.3 ± 0.1	−0.6 ± 0.2	+0.3 ± 0.2

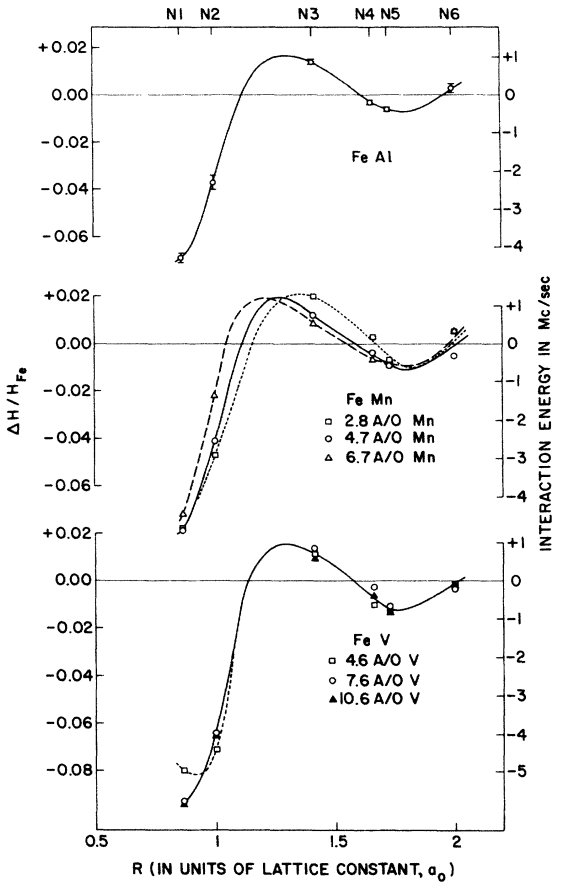

FIG. 2. Variation of conduction-electron spin densities in FeAl, FeMn, and FeV alloys as observed from the internal magnetic field variations as a function of distance from the solute atom. The scale on the right gives the interaction energy in units of Mc/sec. Note that the graph begins at $r = 0.5a_0$, not at the position of the impurity atom.

of the wave vector at the Fermi surface, k_F, is $2.9a_0^{-1}$. This corresponds to a very reasonable value of about 0.4 electrons per atom in these alloys. The experimental and the calculated curves agree excellently for the FeAl series; they give a normalized root mean-square error, g^2, of $(0.2\text{-}0.4)\times 10^{-4}$.

FeMn.—Figure 2 gives the change in the internal field as a function of the distance from a Mn atom. This curve has some different features from that of the FeAl curve. The oscillations are seen to have a variation in phase which probably corresponds to a change in the Fermi radius and band structure. Manifestations of such changes are expected to occur with a solute atom which has its own magnetic structure, as would a transition element. One feature that is, however, similar to the FeAl series is the value obtained for k_F. The similarity of the k_F's is understandable since these alloys are mainly Fe. Excellent fits were obtained for this system, g^2 varying between $(0.2 \text{ and } 0.6)\times 10^{-4}$.

FeV.—Figure 2 gives the FeV results. In general, the fits for this alloy system are poorer than those for the Al and Mn series. We believe this is mainly due to the fact that it seems very difficult to prepare these alloys in a truly random array. In general, the behavior of this series is similar to that of the FeAl system, although some details are different. The spin-density oscillations seem to occur about a value different from H_{Fe}. The value of k_F is again similar to that obtained for the Al and Mn series.

It thus appears that in the FeAl system the core contribution to the internal field remains constant and the observed variation is due to the conduction-electron spin-density behavior.[6] This variation can be compared to similar quantities given in the literature, and upon transformation into wave-vector space yields a direct measurement of the spin susceptibility of the s-conduction electrons. This analysis is discussed in the following Letter.

Further work is in progress to try to obtain the spin-density behavior at larger distances and to obtain more information about the magnetic form factors for a number of systems.

[1]M. A. Ruderman and C. Kittel, Phys. Rev. 96, 99 (1954); K. Yosida, Phys. Rev. 106, 893 (1957).

[2]A. W. Overhauser, Phys. Rev. 128, 1437 (1962).

[3]M. B. Stearns, Phys. Rev. 129, 1136 (1963); Rev. Mod. Phys. 36, 394 (1964); J. Appl. Phys. 35, 1095 (1964).

[4]G. K. Wertheim, V. Jaccarino, J. H. Wernick, and D. N. E. Buchanan, Phys. Rev. Letters 12, 24 (1964).

[5]N. Pessall, K. P. Gupta, C. H. Cheng, and P. A. Beck, to be published.

[6]It is worth noting that the magnitude of the indirect interaction energy measured here seems to correspond directly to that in nonmagnetic materials. Recently C. Froidevaux and M. Weger [Phys. Rev. Letters 12, 123 (1964)], using a spin-echo technique with the Knight shift as a probe, have measured the equivalent quantity to our $N1$ model approximation in nonmagnetic materials. Since our system involves electronic moments whereas theirs involves nuclear moments, we must multiply our results by μ_n/μ_e to obtain the interaction energy we would expect for nonmagnetic alloys. We must also correct for the difference in distance of nearest neighbors in a bcc and a fcc lattice. (Their measurements were on fcc Pt alloys.) If we apply these two corrections to our ΔH_1 values, we would expect an interaction energy for nearest neighbors of about 4 kc/sec in a nonmagnetic fcc system. Their measured values were 4-5 kc/sec which is excellent agreement.