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Localized Versus Itinerant Description of Ferromagnetism in Iron*

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Temperature-dependent hyperfine-field studies of *Fe*Si and *Fe*Mn alloys show that ferromagnetism in iron is a result of long-range exchange interactions, coupling largely localized magnetic moments. This long-range exchange is probably due to the partially itinerant character of the $3d$ electrons.

Studies of hyperfine fields in dilute iron-based alloys at temperatures low compared to the Curie temperature have yielded a considerable amount of information about the spin-density distribution in these alloys. For simple dilutents one can also obtain information about the spin-density distribution in the pure metal, which may lead to a better understanding of the origin of ferromagnetism in iron.

Mössbauer-effect and nuclear-magnetic-resonance spectra of iron alloys consist of a distribution of overlapping peaks which can be attributed to absorbing nuclei having various numbers of impurities in the neighboring shells. An impurity can effect the hyperfine field at neighboring nuclei in several distinct ways: (a) polarization effects produced by the magnetic moment of the impurity causing spin-density oscillations; (b) changes in the magnetic moment of $3d$ character on atoms surrounding the impurity. These changes can occur both in the localized magnetic moment if present or in the spin-density distributions of the itinerant $3d$ electrons. (c) By changing the effective exchange coupling strength of the localized moments of neighboring ions, the temperature dependence of the localized moments and thus the hyperfine field at atoms surrounding the impurity will change.

Until now most of the studies of hyperfine fields in ferromagnetic alloys have been concentrated in the low-temperature range. From these measurements one obtains information about mechanisms (a) and (b). Although various models have been used in attempts to explain these experimental results, it seems quite unlikely that one will be able to determine the relative importance of mechanisms (a) and (b). This is especially so since the more basic problems like the origin of ferromagnetism in iron and the nature of the magnetic moments are as yet unsolved. The most important question really is, "Should the magnetic properties of iron be described using a localized-moment picture or should one use the band description as in the Stoner model?" Perhaps when these problems are solved one will be able to understand the hyperfine fields at low temperatures. A study of the localized versus band description of iron can be done through mechanism (c). To our knowledge the only iron-based alloys on which a detailed temperature-dependent study has been done are the *Fe*Mn alloys.[1] For these alloys the temperature dependence of the Mn^{55} hyperfine field as well as that of Fe^{57} in some of the configurations[2] can be described reasonably well with molecular field theory[2,3] assuming localized moments of $(2-3)\mu_B$ and $2.2\mu_B$ for the Mn and Fe atoms, respectively. In the molecular field theory used, only nearest-neighbor exchange interactions are assumed to be important. Although the localized-moment model cannot explain the results in detail, it does give a good qualitative description of the temperature dependences of the various hyperfine fields. It should be noted that if iron were a pure Stoner ferromagnet, the hy-

perfine fields would be determined by the relative population of the spin-up and spin-down bands. A simple impurity would then only change the relative shift of the spin-up and spin-down bands and the temperature dependence of the hyperfine fields would be independent of the kind of neighboring atoms. The exchange interaction in the band picture is extremely long range so that any disturbance caused by an impurity will be averaged out over the whole crystal. This of course is correct only in the rigid-band picture. In fact the impurity can give rise to local spin- and charge-density disturbances even in the band description. We expect, however, that these local disturbances will not strongly influence the temperature dependences of the hyperfine fields at neighboring ions even though the hyperfine fields at 0°K may be affected considerably.

Therefore, it seems fairly clear that the *Fe*Mn alloys are best described by a localized-moment model with very short-range exchange interactions. By assuming that this is characteristic for the magnetic behavior of the pure host, the temperature dependences of the hyperfine fields at Fe^{57} nuclei in, for example, *Fe*Si alloys are easily calculated since the Fe-Si exchange interaction is zero.

The temperature dependence of the z component of the iron-ion magnetic moment with I silicon impurities in the nearest-neighbor shell is determined by the Brillouin function

$$\langle S_z(I,T)\rangle = SB_s\left(\frac{2J_{ex}S\langle\bar{S}_z(T)\rangle(8-I)}{kT}\right), \qquad (1)$$

where $\langle\bar{S}_z(T)\rangle$ is the z component of the spin averaged over the various configurations: $\langle\bar{S}_z(T)\rangle = \sum_I P(I)\langle S_z(I,T)\rangle$. The calculated magnetization curves for the different neighbor configurations are shown in Fig. 1. $\langle S_z(I,T)\rangle$ decreases more rapidly with increasing T for larger I, and the decreases are more pronounced than the corresponding plots for *Fe*Mn. In the spirit of these considerations we have studied the T dependence of hyperfine magnetic fields in *Fe*Si and, for comparison, also in *Fe*Mn.

The *Fe*Si alloys were prepared by melting together iron and silicon powder in the required proportions. The powders were thoroughly mixed, placed in an Al_2O_3 disk, and melted using a high-frequency furnace. The resulting alloy was rolled to a thickness of 27 μm, then heated at 1100°C, and quenched in water to minimize the degree of order. Mössbauer spectra were recorded in the temperature range from 80 to 1060°K using a

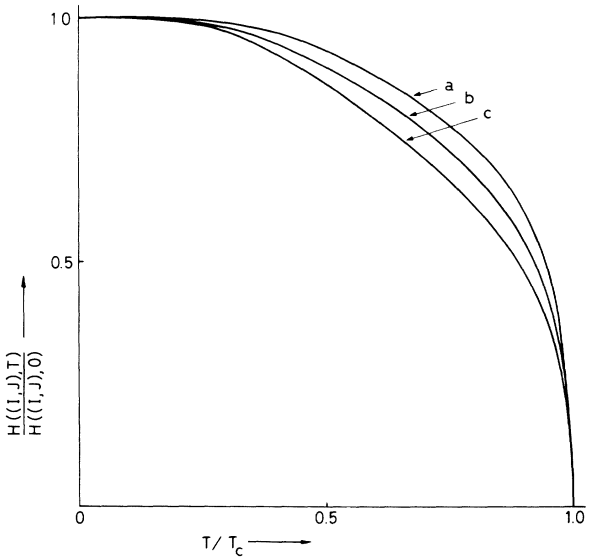

FIG. 1. Calculated magnetization curves a, b, and c for an iron spin with zero, one, and two silicon atoms, respectively, in the first neighboring shell.

cryostat and furnace described in the literature.[4] The temperature of the absorber was constant within ± 0.1°K in the time required to collect a spectrum. The Curie temperature T_c was found using the thermal scanning technique. A typical spectrum of Fe–4 at.% Si obtained at $T = 0.943T_c$ is shown in Fig. 2. The spectra were analyzed assuming only the first two neighboring shells around an iron atom to be important. The probability of finding an iron atom in a particular configuration labeled (I,J) was obtained from the binomial distribution

$$P(I,J) = \binom{8}{I}\binom{6}{J}c^{I+J}(1-c)^{14-I-J}. \qquad (2)$$

Here I and J are the number of Si nearest neighbors in the first and second shells, respectively, and c is the silicon concentration. The spectra were fitted by using the probabilities $P(I,J)$ as constraints and also by constraining the linewidths of all the components corresponding to the same nuclear transition to be equal. It should be noted that we did not constrain the peak positions in any way. It was found necessary to include the outer four lines of the basically six-line Mössbauer spectrum in the fit because of the considerable overlap of lines 1 and 2, and 5 and 6 at elevated temperature. The fit obtained was satisfactory when the configurations (0,0), (0,1), (1,0), (1,1), and (2,0) were included. There was a large uncertainty in the peak position of configuration (1,1), probably a result of some ordering as discussed by Brommer and 't Hooft,[5] and Cranshaw

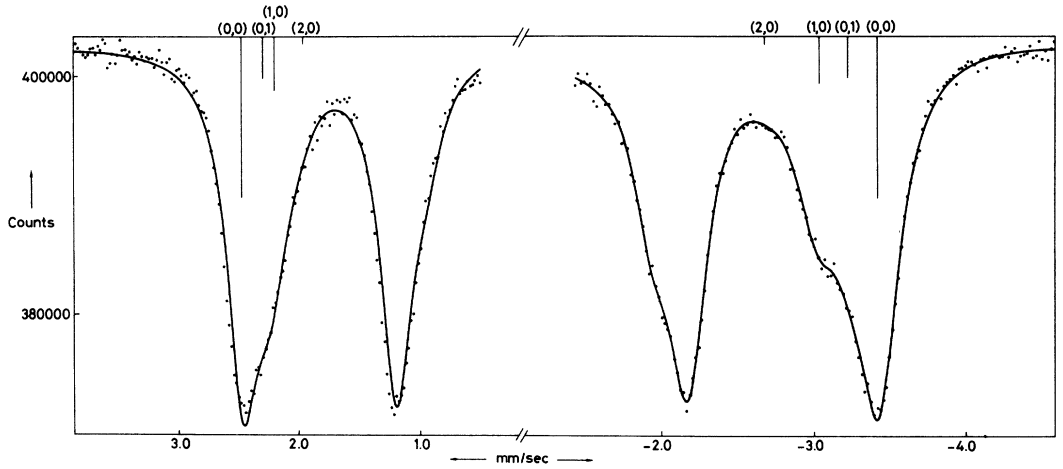

FIG. 2. Outer four peaks of a Mössbauer absorption spectrum of Fe–4 at.% Si at 974.9°K.

et al.[6] Because of the relatively low probability of (1,1), it did not significantly effect the results obtained for the other configurations.

In Fig. 3 are shown the differences of the reduced hyperfine fields,

$$h((I,J),T)=\frac{H((0,0),T)}{H((0,0),0)}-\frac{H((I,J),T)}{H((I,J),0)}. \qquad (3)$$

For comparison we have also shown the results obtained for Fe–3.5 at.% Mn. It is quite obvious that the silicon results do not behave as expected since the spread in $h((I,J),T)$ is much smaller than for Fe–3.5 at.% Mn. As a matter of fact, the temperature dependences of the hyperfine fields for the various configurations are almost the same within the experimental error. The solid curves are obtained from Eq. (1), and the horizontal dotted line for all (I,J) is what we expect for a Stoner ferromagnet in the rigid-band approximation.

Our results obtained for the Fe–4 at.% Si alloy suggest that the exchange interactions in iron are extremely long range, which is quite different from the conclusion arrived at from the Fe–3.5 at.% Mn alloy. It is interesting to note that the Fe–4 at.% Si alloy results can be explained quite well by assuming that ferromagnetism in iron is caused by itinerant electrons as in the Stoner model. Within the rigid-band model we would in this case expect the temperature dependence of the hyperfine field to be independent of the kind of nearest neighbor. Only a very weak direct exchange with neighboring magnetic moments may be present.

Further, it is rather interesting that Si nearest neighbors cause a larger change in the hyperfine

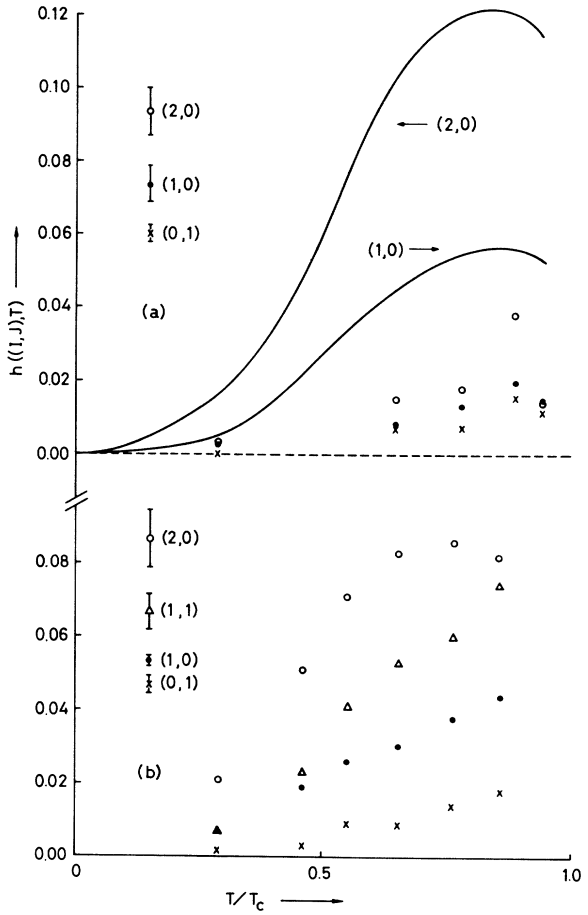

FIG. 3. Differences $h((I,J),T)$ of the reduced hyperfine fields as functions of temperature for (a) Fe–4 at.% Si and (b) Fe–3.5 at.% Mn. The drawn curves in (a) are calculated from expression (1) with one and two silicon atoms, respectively, in the first shell. The dashed line represents $h((I,J),T)$ for all (I,J) for a Stoner ferromagnet in the rigid-band approximation.

field at Fe^{57} nuclei at 0°K than Mn neighbors, while, as first discussed, Mn causes a much larger change in the temperature dependence. This shows that there is no simple relation between the spin-density disturbance caused by an impurity and the exchange interaction. This is in agreement with the conclusion of Stearns[7] who has interpreted the dependence of the hyperfine field on the number of impurity atoms in neighboring shells with the Ruderman-Kittel-Kasuya-Yosida spin-density oscillations, which, however, leads to an antiferromagnetic exchange interaction between nearest-neighbor iron atoms.

In conclusion, (1) the exchange coupling felt by a magnetic moment of a particular iron atom is not locally disturbed by the presence of Si atoms in neighboring shells. Ferromagnetism in metallic iron cannot be explained by a localized-moment model with only short-range exchange interactions. The decrease in T_c of FeSi with increasing Si concentrations shows a decrease in the average exchange coupling as a simple dilution effect. (2) The distribution of hyperfine fields in FeSi alloys is a result of local spin-density disturbances caused by the lack of a magnetic moment on the Si atoms. These local spin-density disturbances, however, have little if any effect on the exchange coupling of the neighboring iron magnetic moments. (3) The different temperature dependences of H_{hf} of Fe^{57} nuclei in different Fe-Mn surroundings show that there is a local variation in the exchange coupling of an iron magnetic moment, which is most likely due to a direct nearest-neighbor Mn-Fe exchange interaction. Such a direct exchange interaction, however, is not characteristic for the pure host.

The experimental results obtained from the FeMn and FeSi alloys lead to the conclusion that iron should be treated as a material with localized magnetic moments which are, however, coupled via long-range exchange interactions. Preliminary calculations based on this model indicate that the experimental results given here can in fact be explained satisfactorily. It is interesting to note that the recent band-structure calculations of Duff and Das[8] yield qualitatively the same physical model for the origin of ferromagnetism in iron. In a subsequent paper we will discuss the results in terms of this model.

Similar studies of the iron based alloys containing Al, V, Cr, Co, and Ni are in progress.

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Spin Susceptibility of Disordered Binary Alloys*

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A new theory based on the coherent-potential approximation is presented for the spin susceptibility χ in disordered binary alloys having arbitrary concentration and scattering-potential strengths. In the dilute-alloy limit the susceptibility contains all formerly derived expressions for χ as well as new physically significant terms. In the weak-scattering limit, an extension of the local-exchange-enhancement model is obtained which can be satisfactorily applied to nondilute Pd-Pt.

There exist a number of magnetic or nearly magnetic disordered alloys A_xB_{1-x} which form solid solutions over a wide range of concentrations. Among these are Ni-Rh, Ni-Pd, Rh-Pd, and Pt-Pd. All of these systems show interesting magnetic behavior in the nondilute alloy limits. For example, Ni-Rh becomes ferromagnetic[1] at 63 at.% Ni; the spin susceptibility of Ni-Pd increases nonlinearly with Ni concentration[2] below 2 at.%; in Rh-Pd alloys, χ exhibits a max-