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Magnetization Distribution in Magnetically Diluted Systems with Application to Iron-Palladium*

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In magnetic systems that have been substitutionally diluted there exists a large number of different magnetic sites the diversity of which is determined by the nature of the magnetic interaction and the statistical fluctuations in the distribution of the magnetic atoms. We have calculated the magnetic and exchange field distributions from the magnetic atom concentration, the lattice structure, and the magnetic interaction potential in a self-consistent way for any temperature below the transition temperature. The calculation has been applied to the case of the hyperfine field distributions observed by nuclear magnetic resonance and by the Mössbauer effect in magnetic random substitutional alloys where the interaction is long-range.

INTRODUCTION

CENTRAL to the problem of metallic magnetism is an understanding of the existence of magnetic impurities in nonmagnetic hosts and their interactions. These two questions cannot be considered independently, for it is probable that the interactions may, in fact, stabilize the existence of the localized magnetic state.[1] Unfortunately, it is not possible to determine the nature of the interactions directly. However, under certain assumptions which can be verified experimentally,[2] it is possible to relate the distribution of magnetic hyperfine fields of the impurity or host atoms to the impurity-impurity interactions.

Most experimental investigations[3–6] have focused attention on the region where the magnetization is saturated and have investigated the effect of impurities on the hyperfine field. In contrast, this paper is concerned with the relationship between the distribution of hyperfine fields and the magnetic interactions in the region where the magnetization is not saturated. The view taken here is that each magnetic atom produces an exchange field proportional to the polarization of that atom. Each magnetic atom aligns itself in the exchange field at its site. Statistical fluctuations in the density of the magnetic atoms produce fluctuations in the exchange field which in turn produce a distribution in the magnetization. This distribution is generally broad near the transition temperature and narrow at low temperatures as all the magnetic atoms become completely polarized.

When the interaction is short-range only the first few sets of nearest neighbors are involved and the number of significant moment configurations is small. In this case the atoms contributing to the exchange field are small in number and may be directly summed. In contrast, this paper will be concerned with long-range interactions and the attendant problem of numerous moment configurations.

Woodhams, Meads, and Carlow[7] have also suggested this mechanism as the cause of the observed hyperfine field distribution in iron-palladium alloys. Dunlap and Dash[8] have taken a similar view and have determined the exchange energy and range parameter by applying a cell model to the Mössbauer data on CoPd alloys. Takahashi and Shimizu[9] have considered the same problem using an itinerant electron model which does not take into account the magnetization distribution. In a preliminary Monte Carlo approach[2] to this problem, the distribution of hyperfine fields in the ferromagnetic dilute FePd alloys was shown to be consistent with a random distribution of iron atoms and an exchange field proportional to the electron polarization determined by diffuse neutron scattering from FePd alloys.[10] This work also presented some evidence of short-range metallurgical ordering and discussed the shortcomings of the Monte Carlo calculation, primarily the lack of self-consistency.

In the first section of the present work a general calculation of the probability distribution of the exchange field $P(J)$ is made, an analytic calculation

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is performed for a few restricted cases, and the results are compared with previous work. In the second section the general calculation is expanded to achieve self-consistency and to account for short-range order. In the third section the $P(J)$ for dilute FePd alloys is determined and the results are compared with the earlier Monte Carlo calculations. The calculation is done by assuming that the exchange field about an Fe atom is Gaussian and determining the width of the Gaussian by the χ^2 test from the Mössbauer hyperfine field spectra. From this $P(J)$, as determined from the Mössbauer experiments, the low-temperature iron NMR spectra and the average magnetization are calculated. These are compared with experimental results. The expected NMR spectra of palladium are also calculated but do not agree with the experimental results for any simple assumptions. In the final section the assumptions and results of this work will be compared with those of other approaches to this and similar dilute alloy problems.

FORMULATION OF $P(\mathbf{J})$

Consider the simple case of a random substitutional alloy composed of two types of atoms, one of which is magnetic and the other is not. If a magnetic atom is selected at random, what is the probability $P(\mathbf{J})$ that this atom experiences an exchange field $\mathbf{J}$?[11] This question will be investigated under the following assumptions:

(1) The exchange field $\mathbf{J}$ at a particular atomic site is the vector sum of the exchange fields originating from the neighboring magnetic atoms.

Consider that there are various classes of neighboring atomic sites which when occupied with a magnetic atom produce a contribution $\mathbf{v}_k$ at the given site. The kth class has m_k sites and contains n_k magnetic atoms, and this class provides a contribution $n_k\mathbf{v}_k$ at the given site. The probability $P(\mathbf{J})$ is then given by

$$P(\mathbf{J}) = \sum_{\{n_k\}}\prod_k P_k(n_k)\,\delta(\mathbf{J}-\sum_k n_k\mathbf{v}_k), \tag{1}$$

where $P_k(n_k)$ is the probability that the kth class of sites contains n_k magnetic atoms, the primary sum is over all possible configurations n_k, and $\delta(x)$ is the Dirac δ function. If the alloy is a random substitutional alloy,

$$P_k(n_k) = m_k!\,c^{n_k}(1-c)^{(m_k-n_k)}/n_k!\,(m_k-n_k)!, \tag{2}$$

where c is the fraction of atoms which are magnetic. Using the Fourier integral representation of the δ function,

$$\delta(\mathbf{J}-\sum_j n_j\mathbf{v}_j) = \frac{1}{(2\pi)^3} \times \int_{-\infty}^{\infty}\exp(i\boldsymbol{\rho}\cdot\mathbf{J})\prod_j \exp(-in_j\boldsymbol{\rho}\cdot\mathbf{v}_j)\,d\boldsymbol{\rho}, \tag{3}$$

Eq. (1) becomes

$$P(\mathbf{J}) = \frac{1}{(2\pi)^3}\int_{-\infty}^{\infty}\exp(i\boldsymbol{\rho}\cdot\mathbf{J}) \times\prod_k\sum_{n_k}P_k(n_k)\exp(-in_k\boldsymbol{\rho}\cdot\mathbf{v}_k)\,d\boldsymbol{\rho}. \tag{4}$$

$P_k(n_k)$ is the n_kth term of the binomial expansion of $[(1-c)+c]^{m_k}$, and similarly $P_k(n_k)\exp(-in_k\boldsymbol{\rho}\cdot\mathbf{v}_k)$ is the n_kth term of $A_k=[(1-c)+c\exp(-i\boldsymbol{\rho}\cdot\mathbf{v}_k)]^{m_k}$. From this

$$P(\mathbf{J}) = \frac{1}{(2\pi)^3}\int_{-\infty}^{\infty}\exp(i\boldsymbol{\rho}\cdot\mathbf{J}) \times\prod_k\{1-c[1-\exp(-i\boldsymbol{\rho}\cdot\mathbf{v}_k)]\}^{m_k}d\boldsymbol{\rho}, \tag{5}$$

an exact result.

It is useful to write

$$A_k = \exp(\ln A_k),$$

which for $c\ll 1$ becomes

$$A_k \cong \exp\{-cm_k[1-\exp(-i\boldsymbol{\rho}\cdot\mathbf{v}_k)] \times[1+c[1-\exp(-i\boldsymbol{\rho}\cdot\mathbf{v}_k)]/2]\}. \tag{6}$$

By retaining only the exponential term linear in c, the binomial distribution $P_k(n_k)$ has been approximated by the Poisson distribution.

It should be noted that the case $c\ll 1$ is not necessarily the low-concentration limit of only a few interacting magnetic atoms since the range of the interaction may be quite large. Nevertheless the expansion is valid.

(2) The exchange field at a particular atomic site is parallel to the exchange field at any other site.

Thus $\mathbf{v}_n$ and $\boldsymbol{\rho}$ become scalar quantities and the normalizing factor in the Fourier transform goes to $1/2\pi$. The effect of this Ising approximation is to impose a strong correlation on the z component of the exchange field while ignoring correlations in the x and y components. This assumption is not unreasonable for simple ferromagnetic and strict antiferromagnetic alloys not too near the transition temperature.

(3) The v_j are real.

Using the Poisson approximation

$$P(J)\cong\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp[-c\sum_k m_k(1-\cos\rho v_k)] \times\cos(\rho J-\sum_k cm_k\sin\rho v_k)\,d\rho, \tag{7}$$

a rather simple result. An even simpler result holds for random spin orientation. In this case for every class k there is a class k' such that $m_{k'}=m_k$ and $v_{k'}=-v_k$, so

$$P(J)\cong\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp[-c\sum_k m_k(1-\cos\rho v_k)] \times\cos(\rho J)\,d\rho. \tag{7'}$$

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[11] T. A. Kitchens and W. L. Trousdale, J. Appl. Phys. **39**, 1321 (1968).

This equation is analogous with Eq. (2.8) of Klein and Brout.[12] Equations (7) and (7′) yield nearly the same result at the Monte Carlo calculation presented earlier.[2]

Now consider the sum in the exponential in Eq. (7), each term of which is positive. The sum is always nonzero unless $\rho=0$ or ρ takes on a value ρ_0 such that $\rho_0 V_k=0$ (mod2π) for all k. This second condition is unlikely in any realistic problem, so the major contribution to the integral arises from small ρ, especially if the m_k are large or numerous. Thus, for long-range interactions where $c\sum_k m_k\ (1-\cos\rho v_k)$ is large for large ρ, the trigonometric functions of ρV_k may be expanded about $\rho=0$. Equation (7) becomes

$$P(J)\cong\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp(-\rho^2 c\sum_k m_k v_k^2/2)$$

$$\times\cos[\rho(J-c\sum_k m_k v_k)]d\rho$$

$$\cong[1/(2\pi)^{1/2}\sigma]\exp[-(J-\bar{J})^2/2\sigma^2]. \tag{8}$$

This result is exactly the intuitive answer; the distribution is Gaussian with $\bar{J}=c\sum_k m_k v_k$, the average exchange field, and $\sigma^2=c\sum_k m_k v_k^2$, the square of the standard deviation. For random spin orientation $\bar{J}=0$, as explained above.

Although the present result is Gaussian we see $P(J=0)\propto 1/\sqrt{c}$ rather than $1/c$, the dependence suggested by Marshall[13] and calculated by Klein and Brout[12] for the RKKY[14–16] interaction where $\bar{J}=0$. The difference arises because the interaction is assumed to be sufficiently long-range that even for small c many atoms contribute to the exchange field at the origin. This means that, despite the small c, this analytic result is still in the "high-concentration region" discussed by Anderson[17] and by Kittel and Abrahams.[18]

SELF-CONSISTENCY AND SHORT-RANGE ORDER EFFECTS

The calculation in the previous section was not done in a self-consistent way. Previously the exchange field distribution at the origin has been calculated assuming that each magnetic atom made a unique contribution to the exchange field at the origin. In this section, self-consistency will be introduced by assuming that every atom has the same probability distribution as the one at the origin, $P(J)$.

Also in this section we will account for metallurgical or chemical spatial short-range order. There is some evidence for such ordering in metallurgical solid solutions such as FePd.[2] Magnetic correlations, i.e., short-range order which depends on the polarization of the atom at the origin, will be considered in the discussion.

In order to account for the effects of self-consistency and short-range order we expand on the formulation given in the last section. Equation (1) can be written as

$$P(J)=\prod_j\sum_{n_j}[P_j(n_j)\delta(\mathbf{J}_j-n_j\mathbf{v}_j)]\delta(\mathbf{J}-\sum_j\mathbf{J}_j), \tag{9}$$

which can be generalized to

$$P(\mathbf{J})=\prod_j\sum_{n_j}[P_j(n_j)\prod_{i=0}^{n_j}\int P_{ji}'(\mathbf{v}_{ji})$$

$$\times\delta(\mathbf{J}_j-\sum_{i=0}^{n_j}\mathbf{v}_{ji})d\mathbf{v}_{ji}]\delta(\mathbf{J}-\sum_j\mathbf{J}_j), \tag{10}$$

where $P_{ji}'(\mathbf{v}_{ji})$ is the probability that the ith magnetic atom placed in the jth class produces a contribution $\mathbf{v}_{ji}$ to the exchange field at the origin. Let $P_{j0}'(\mathbf{v}_{j0})=\delta(\mathbf{v}_{j0})$, $P_{ji}'(\mathbf{v}_{ji})=P_j'(\mathbf{v}_{ji})$, and $\mathbf{v}_j=\mathbf{v}_{ji}$. These restrictions mean that all spatial ordering is accounted for in $P_j(n_k)$ and that the probability that a magnetic atom produces a certain contribution to the exchange field is not dependent on the number of magnetic atoms already in that class. We replace $\delta(\mathbf{J}-\sum_j\mathbf{J}_j)$ with its Fourier representation:

$$P(\mathbf{J})=\frac{1}{(2\pi)^3}\int\exp(i\boldsymbol{\varrho}\cdot\mathbf{J})\prod_j\sum_{n_j}P_j(n_j)\psi_j(\boldsymbol{\varrho})^{n_j}d\boldsymbol{\varrho}, \tag{11}$$

where

$$\psi_j(\boldsymbol{\varrho})=\int\exp(-i\boldsymbol{\varrho}\cdot\mathbf{v}_j')P_j'(\mathbf{v}_j')d\mathbf{v}_j'. \tag{12}$$

By looking at Eqs. (4) and (11) it is easy to see that $\psi_j(\boldsymbol{\varrho})$ has replaced $\exp(-i\boldsymbol{\varrho}\cdot\mathbf{v}_j)$. Thus the equations assuming random substitutional alloys in the previous section can be made self-consistent by making this substitution.

Finally the polarization of any atom is assumed to be proportional to the Brillouin function, $B_S(J/kT)$, for an atom with spin S in an exchange field J at temperature T. In this case

$$P_j'(\mathbf{v}_j')=\int P(\mathbf{J})\delta(\mathbf{v}_j'-\mathbf{v}_j^0 B_S(J/kT))d\mathbf{J}, \tag{13}$$

where $\mathbf{v}_j^0$ is a constant vector collinear with $\mathbf{J}$.

APPLICATION TO DILUTE FePd ALLOYS

In this section the self-consistent formulation is used to calculate the hyperfine field in dilute FePd alloys. This is accomplished by assuming that the spatial shape of the exchange field produced by an Fe atom is an isotropic Gaussian. The width of the Gaussian and a scaling parameter J_0 are found by the method of least squares (χ^2 test) from the Mössbauer hyperfine spectra at a given temperature T.

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The predicted spectra for various temperatures are found to agree with the experimental spectra except near the ordering temperature. This behavior is, however, expected because correlations in the motions of the moments are expected to have a large effect near the ordering temperature and correlations have been neglected. The $P(J)$ derived in the above calculation is then used to predict the average or bulk magnetization in dilute FePd alloys and comparison is made with experimental data. The $P(J)$ is also used to calculate the nuclear magnetic resonance line shape for the Fe and also for the Pd and these predictions are compared with the data of Budnick *et al.*[19]

Again, as in the earlier Monte Carlo calculation[2] the exchange field surrounding the magnetic Fe atom is assumed to be carried by the electron polarization which has been observed by neutron scattering to be almost Gaussian, i.e., $p\sim\exp(-r^2/2\sigma^2)$.[10] Thus the physical picture used is a localized model where the localized moment is of large spatial extent due to the high polarizability of the Pd host. Simple band theory would suggest a uniform polarization of the Pd host and this uniform polarization would not be observed by the neutron scattering experiment. Using the localized model, the exchange is proportional to the three-dimensional overlap integral or

$$v_k^0 = J_0 \exp(-r_k^2/\sigma^2), \tag{14}$$

where r_k is the distance to the kth-nearest-neighbor class, and σ is the width of the electronic polarization. We account explicitly for the first 16 nearest-neighbor sites and then take mean r_k's for groups of 50, 100, 150, etc., atoms until 95% of the contribution to the average exchange field is included.

In order to illustrate the effect of self-consistency we have assumed the alloys to be random substitutional and have calculated $P(J)$ by iterating Eqs. (11)-(13), where

$$\sum_{n_j} P_j(n_j)\psi_j(\rho) = \{1-c[1-\psi_i(\rho)]\}^{m_j}$$

was approximated by its expansion to second order in c. Once $P(J)$ is determined we can calculate the distribution of the hyperfine field $D(H)$ by assuming that the hyperfine field H is proportional to the polarization of the atom,

$$D(H) = \int_{-\infty}^{\infty} P(J)\,\delta(H - H_{\mathrm{sat}} B_s(J/kT))\,dJ, \tag{15}$$

where H_{sat} is the experimentally determined saturation hyperfine field and the spin $S=3.5$, in agreement with the experimentally determined value.[20] It has been shown earlier in this laboratory that the average hyperfine field is proportional to the bulk or average magnetization for a particular dilute FePd alloy, $\mathrm{Fe}_{2.65}\mathrm{Pd}_{97.35}$.[21]

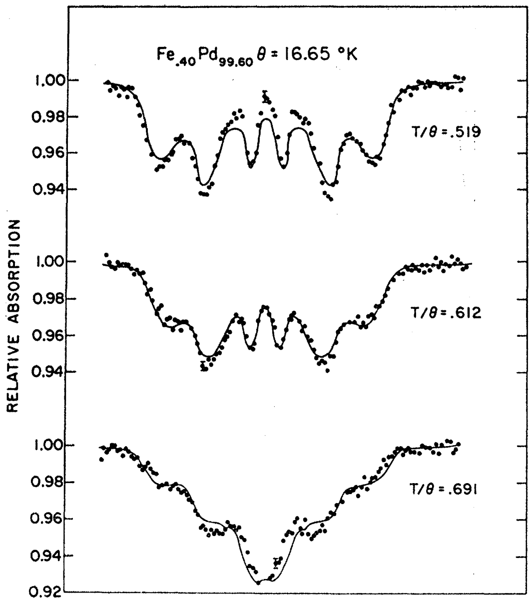

FIG. 1. The experimental and calculated Mössbauer spectra for $\mathrm{Fe}_{0.40}\mathrm{Pd}_{99.60}$ at various reduced temperatures.

Mössbauer spectra can be calculated from this distribution function by

$$M(v) = \int_{-\infty}^{\infty} D(H) \sum_{i=1,6} L_i(H - \alpha_i v)\,dH, \tag{16}$$

where L_i is the line function, a Lorentzian for ideally thin source and absorbers, the α_i are known from the nuclear excited- and ground-state magnetic moments for Fe^{57}, and v is the velocity. In this calculation there is an implicit assumption that there is a distribution of ferromagnetic domains sufficient in size and number to justify the Ising-model assumption and the summation over all possible configurations in $P(J)$ in every direction.

Mössbauer data for $\mathrm{Fe}_{0.40}\mathrm{Pd}_{99.60}$ was used to determine the two free variables of this calculation, J_0 and σ, by the χ^2 test. The experimental spectra and calculated spectra for the best values of J_0 and σ are shown in Fig. 1. The exchange distributions corresponding to the calculated spectra are shown in Fig. 2. The best J_0 and σ are plotted in Fig. 3 as a function of the reduced temperature, T/θ, and the results from the Monte Carlo calculations are shown for comparison.

The χ^2 of the new calculation of the Mössbauer spectra is not significantly better than the Monte Carlo calculation. It is reassuring that the calcula-

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[19] We are grateful to J. I. Budnick and co-workers for unpublished data on the temperature dependence of the palladium resonance in $\mathrm{Fe}_{0.05}\mathrm{Pd}_{99.5}$.

[20] M. P. Maley, R. D. Taylor, and J. L. Thompson, J. Appl. Phys. **38**, 1249 (1967).

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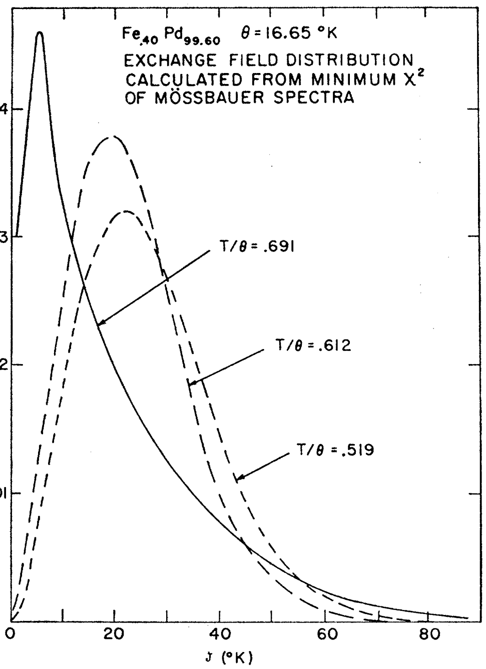

FIG. 2. The exchange field distributions $P(J)$ for the same reduced temperatures used in Fig. 1.

tions agree at low temperatures where the effects of self-consistency are less important and that the new σ and J_0 remain constant to higher temperatures than did the values calculated from the non-self-consistent Monte Carlo method. The values of σ at $T/\theta<0.7$ are in excellent agreement with the σ determined by neutron scattering for the electron polarization. This strongly supports the basic assumptions made in the calculation. The deviations are, however, severe at $T/\theta>0.7$, indicating that effects of both static and dynamic correlations are important. The decrease of σ as T approaches T_c was discussed previously.[2]

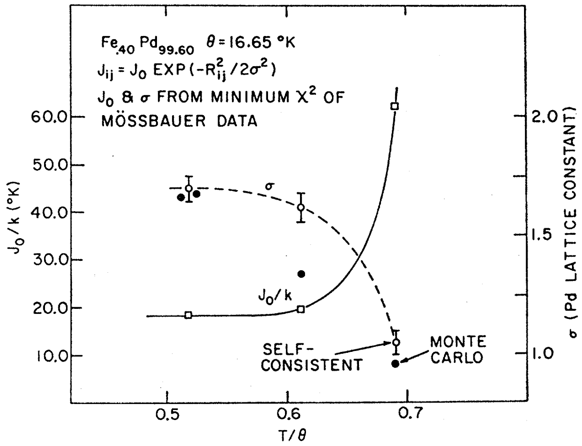

FIG. 3. The exchange constant and range parameter as a function of reduced temperature for $Fe_{0.40}Pd_{99.60}$.

In Fig. 4 is presented the reduced average hyperfine field $\langle H\rangle/H_{\text{sat}}$, which has been shown to be equal to the reduced average magnetization $\langle M\rangle/M_{\text{sat}}$ as calculated from the low-temperature values of J_0 and σ derived from the $Fe_{0.40}Pd_{99.60}$ Mössbauer data. This calculation predicts a critical temperature $\theta=14.4$°K rather than 16.6°K, a discrepancy expected from the fact that within the model J_0 and σ are not independent of T. (See Fig. 3.) For comparison the reduced average magnetization from the molecular-field model for $S=3.5$ is shown. Magnetization measurements by Crangle[22] for 1.25, 3.15, and 5.11% Fe, Mössbauer hyperfine field measurements for 2.65% Fe[20] and for 0.50% Fe,[2] and average NMR hyperfine fields[23] for 0.50% Fe are also shown in Fig. 4. Within the experimental accuracy higher concentrations tend toward the molecular-field result as expected. The Craig *et al.*[21] Mössbauer data are high because they have measured the most probable hyperfine field rather than the average hyperfine field. This is expected as shown by the NMR results and correctly predicted by the model as shown in Fig. 6.

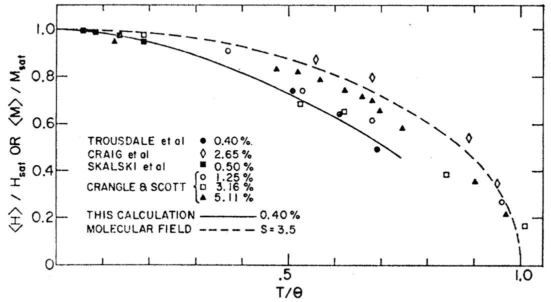

FIG. 4. The reduced magnetization (or hyperfine field) versus the reduced temperature for dilute FePd alloys from this calculation and for the molecular-field approximation (dashed line). Mössbauer data for 0.40 and 2.65% Fe, magnetization data for 1.25, 3.16, and 5.11% Fe, and NMR data for 0.50% Fe are also shown.

Nuclear magnetic resonance for Fe^{57} in $Fe_{0.50}Pd_{99.50}$ has been observed by Skalski *et al.*[23] at low T/θ. They have observed that the hyperfine field varies as $T^{3/2}$, behavior characteristic of spin waves. Although the present model has neglected correlations such as spin waves, the average magnetization varies as T^n where n is approximately 2 over the extraordinarily long region of $T/\theta\lesssim0.5$. Their average hyperfine field measurements are compared with the model calculation on a reduced temperature scale assuming $\theta=22.0$°K for their sample and their extrapolated value of $H_{\text{sat}}=-301.2$ kOe, in Fig. 5. The scaling of the NMR data depends in a rather sensitive way on the choice of θ. The transition temperature was not measured for this sample but a choice of 22°K does not seem unreasonable based on a determination of $\theta=$

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23°K by Crangle and Scott[24] from susceptibility for $Fe_{0.53}Pd_{99.47}$. This determination of the transition temperature is based on an extrapolation method which has led to larger values of θ than that measured by the Mössbauer method. It should be noted that the low-temperature NMR data on Fe and our calculation are in excellent agreement.

In addition to the magnetization versus temperature as measured by NMR it is also possible to calculate the line shapes of the NMR resonance. Under the assumption that instrumental line broadening is small compared with observed linewidths, the NMR line shape for the iron atoms and the probability distribution of fields $[D(H)]$ are the same. It is known, however, that the average saturated hyperfine field H_{sat} is dependent on the average concentration in dilute FePd alloys,[25] and this implies that H_{sat} is a function of the local environment. Evidence for this is seen in the low-temperature NMR data illustrated at the top of Fig. 6. The present model has assumed that H_{sat} is independent of the local environment. It therefore predicts a line that is narrower but of the correct shape. If the predicted line were folded into the low-temperature NMR data, it would agree well with the observed NMR line profiles. Furthermore spin-wave excitations have been neglected in this calculation. These excitations are expected to affect the regions of highest magnetic order to a greater degree than regions of low magnetic order. For this reason the most probable value of the $D(H)$ will occur at lower H but the average H will not be so strongly affected.

This model does not predict the position and shape of the Pd NMR line without further assumptions. The simplest postulate is that the palladium resonance is linearly dependent on the local electron polarization ρ. In this case it is implicitly assumed that any p- or d-electron polarization would only amplify the s-electron polarization in a linear manner. The probability distribution of electron polarization $P(\rho)$ can be calculated in the same way as $P(J)$ from Eqs. (11), (12), and (13), except that v_j^0 is replaced by $\rho_0 \exp(-r_j^2/2\sigma^2)$. This comparison has been made with NMR data for $Fe_{0.5}Pd_{99.5}$ [19] at $T=4.2$ and 3.25°K. The asymmetry of the NMR line and its temperature independence at low temperatures is correctly predicated but the experimental linewidth is three times narrower than the one calculated by this model.

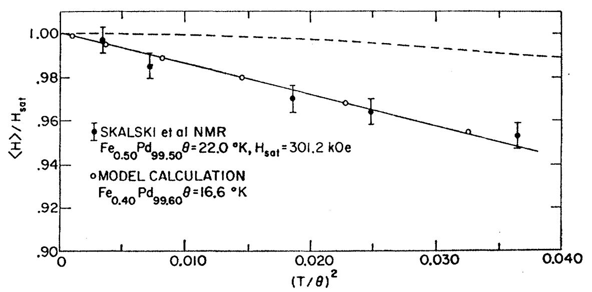

FIG. 5. The low-temperature reduced hyperfine field from this model and NMR data. The model calculation dependence for $Fe_{0.50}Pd_{99.50}$ has been checked at $(T/\theta)^2=0.0037$ and agrees with that calculated for $Fe_{0.40}Pd_{99.60}$.

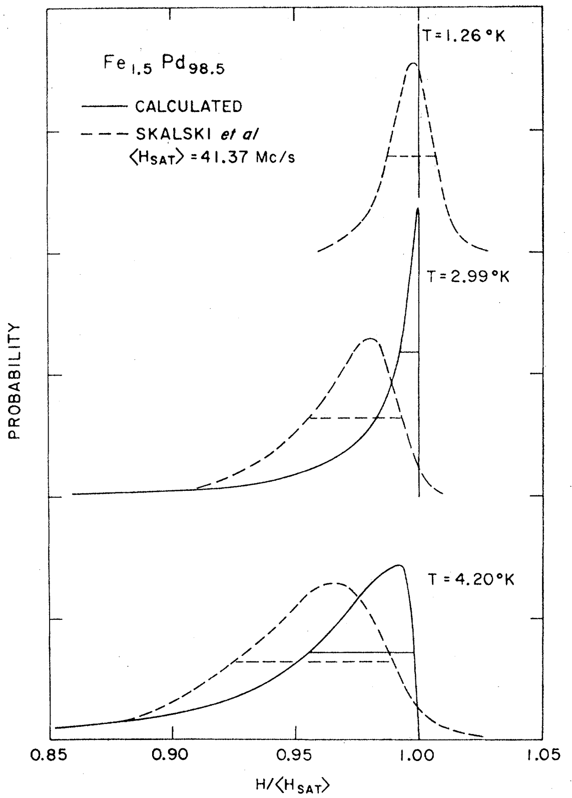

FIG. 6. The calculated and experimental NMR spectra for $Fe_{1.5}Pd_{98.5}$. The low-temperature J_0 and σ found by the χ^2 test for $Fe_{0.40}Pd_{99.60}$ were used in the model calculation.

A second simple postulate is that each palladium atom has an associated moment whose value is independent of the local environment. This moment could, for instance, arise from the changes in electronic band structure due to alloying, since local moments appear to be absent in pure palladium. The alignment of these moments could be calculated by assuming, for example, a local polarization field as above. Now either (1) the Pd moment is small and is not saturated at $T<\theta$, or (2) the Pd moment is large and saturated at $T<\theta$. In the first case the NMR line would have the same asymmetry as the $P(J)$ curves but would be temperature-dependent. In the second case the NMR line would have the asymmetry of the Fe NMR lines and would be temperature-independent. Since the NMR lines have the $P(J)$ asymmetry but are temperature-independent, the postulate of an environment-independent moment is not correct.

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A third possibility is that in addition to the $P(J)$ that we have calculated there is a uniform polarization of the band which is principally responsible for the palladium resonance. $P(J)$ then produces a broadening of this resonance. Again in view of the excellent agreement with the magnetization, NMR on iron transition temperatures, and Mössbauer spectra from the calculated $P(J)$, this postulate seems unlikely. In reality there are probably moments induced on the Pd atoms dependent upon the local environment as has been suggested by Skalski *et al.*[23]

The specific heat of iron-palladium alloys has been measured by Veal and Rayne.[26] We have not attempted to fit this calculation to their data since the major portion of the contribution to the specific heat comes in the region near θ where the calculation is not valid, as discussed above.

SUMMARY AND CONCLUSIONS

A phenomenological theory of the probability distribution of local magnetization of dilute magnetic alloys has been developed which can be regarded as two steps closer to reality than the molecular-field approximation. The theory has been applied to the dilute FePd alloys assuming the magnetic interaction is an overlap of the host polarization localized about the Fe atoms. This polarization is assumed to be Gaussian in spatial extent $[\rho\sim \exp(-r^2/2\sigma^2)]$. From applying the χ^2 test to Mössbauer magnetization distribution measurements σ is determined and found to agree with σ determined by diffuse magnetic neutron scattering. This model neglects the effects of itinerant electrons except in the sense that the magnetic interaction is assumed to be an RKKY interaction which has been enhanced so strongly as not to oscillate in sign until it is weak enough to be neglected. Since dynamic fluctuations, a subject discussed in a more elegant theory by Doniach and Wohlfarth,[27] have been neglected, it was expected that the predictions made from this model were poor for temperatures greater than 70% of the ordering temperature. For this reason no meaningful comparison could be made to the specific heat. It was found that, under some simple assumptions, this model could not predict the correct line shapes or temperature dependence for hyperfine fields of palladium atoms in dilute FePd alloys. Some explanation of these difficulties will, no doubt, come from an understanding of the nearly ferromagnetic palladium host. This question is discussed in some recent work of Doniach and Murani[28] and also by Shimizu and Takahashi.[29] Both of these groups have given an explanation of how the Mössbauer and susceptibility data can suggest a spin value $\gtrsim 3.5$,[20] while the specific-heat data are consistent for a spin value of less than $\frac{3}{2}$.[26] The exact spin value does not change the present calculations in any significant way.[11]

On the other hand, this model has been successful in explaining both the line shape and the temperature dependence of the NMR and the Mössbauer hyperfine spectra for the iron atoms in dilute FePd alloys. The model has also predicted, with surprising success, the temperature dependence of the average hyperfine field at low temperatures with no additional free parameters.

Klein and Brout[12] have also calculated the $P(J)$ in random magnetically dilute systems for the case of oscillatory RKKY interactions. This work can be regarded as complementary to the present approach since the effects of correlations have been emphasized while the combinatorial problem has been avoided by considering only the very dilute case. Klein[30] has worked on the difficult task of extending this approach to nonzero temperatures and has predicted the very-low-temperature properties. The general features of systems such as dilute MnCu which have no discrete ordering temperature are in a qualitative agreement with these calculations.

It is clear that our general approach could be extended to such systems and the temperature dependence enters in a reasonable and direct way. Very recently Klein[31] has, in fact, independently developed the approach that was used in this work and has specialized it to oscillating exchange fields and very dilute concentrations. With this specialization Klein has again neglected the combinatorial problem and has found analytic expressions for the high- and low-temperature values of the magnetic susceptibility, the specific heat, and the width of the $P(J)$ distribution. This work also illustrates the power of this formulation, which is an extension of the Bethe-Peierls-Weiss method but does not have the "anti-Curie-point" difficulties.[32] It is anticipated that the method will be of significant value in leading to an understanding of the nature of the magnetic ordering process in a number of materials.

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