

Model of localized moments for dilute *PdFe* alloys

R. Medina and R. E. Parra

*Centro de Física, Instituto Venezolano de Investigaciones Científicas, Apartado 1827,
Caracas 1010A, Venezuela*

(Received 8 June 1982)

A model that describes several magnetic properties of *PdFe* alloys with concentrations of $0.03 < c < 0.5$ at. % Fe is presented. An expression is derived for the magnetic-moment distribution of the magnetization cloud which gives results that agree with experimental data. An expression for the moment-dependent part of the energy is obtained. From this expression an effective Heisenberg interaction between magnetization clouds is derived. The stiffness constant of spin waves is calculated and found consistent with experimental results. With the use of the Monte Carlo method for a classical Heisenberg system, the critical temperatures and the magnetic susceptibilities are estimated and show good agreement with experimental data at different concentrations. The model supports the idea of polarization clouds produced by first-neighbor interactions between moments.

I. INTRODUCTION

Alloys of *PdFe* have long been considered typical examples of giant-moment ferromagnetic systems. Magnetization data^{1,2} and neutron-diffraction measurements^{3,4} give a $10\mu_B$ moment per iron atom. Most of this giant moment resides in the palladium atoms, which form a polarization cloud around each iron atom with an extent of $\sim 10 \text{ \AA}$, as has been measured with neutrons by Low and Holden.³ At concentrations as low as 0.1 at. % Fe, the magnetization clouds overlap making the alloy ferromagnetic at low temperatures.

Doniach and Wohlfarth⁵ developed a theory for dilute *PdFe* alloys in which the palladium *d* holes are polarized by the iron moments through a nonlocal exchange-enhanced susceptibility. Zuckermann⁶ extended the theory in order to include local exchange effects. These theories did not yield the shape of the magnetization cloud, and the predicted value of the spin-wave stiffness coefficient *D* was much smaller than the experimental one.⁶

Recently we developed a model for Ni-Pd and Ni-Pt alloys⁷ which successfully described the static magnetic properties of these systems. When this model was applied to *PdFe* it correctly predicted the range of the polarization clouds.

Our theory, in the *PdFe* case, reduces to a model where local palladium moments are proportional to the moment of its neighbors. Linear-response theory is justified by the value of the Pd moment, $0.07\mu_B$ for a first neighbor of an Fe atom, which is small in comparison with the estimated number of

holes per atom (0.36). In contrast to the nonlocal susceptibility model, the Fe moment polarizes directly only its first neighbors.

In this paper we extend our previous model in order to include an expression for the energy which allows us to reduce the interaction between magnetization clouds to a Heisenberg form.

II. MAGNETIZATION CLOUDS

The model we apply to *PdFe* assumes that the Fe moments are insensitive to their local magnetic environment while the moment of a Pd atom at site \vec{n} is given by

$$\mu_{\vec{n}} = \chi_0 h_{\vec{n}}, \quad (1)$$

where χ_0 is the nonenhanced susceptibility and $h_{\vec{n}}$ is a local field given by

$$h_{\vec{n}} = b_{\vec{n}} + J \sum_{\vec{\delta}} \mu_{\vec{n} + \vec{\delta}} [1 + (\alpha - 1) p_{\vec{n} + \vec{\delta}}]. \quad (2)$$

Here $b_{\vec{n}}$ is the external field, $p_{\vec{n}}$ is a site occupation operator which is equal to zero if a Pd atom is at \vec{n} and equal to one otherwise, $\mu_{\vec{n} + \vec{\delta}}$ is the magnetic moment of a first neighbor, *J* is the exchange constant between Pd atoms, and αJ is the exchange constant between Fe and Pd.

In the case of pure Pd in a homogeneous external field, Eq. (2) implies that each Pd atom "feels" an additional field of value $J\mu$ from each of its *Z* first

neighbors. The actual susceptibility is then, not χ_0 , but the following enhanced susceptibility:

$$\chi = \frac{\chi_0}{1-\Gamma}, \quad (3)$$

where the magnetic enhancement factor Γ is given by

$$\Gamma = JZ_1\chi_0. \quad (4)$$

We may use this model to calculate the magnetization cloud produced by a single Fe atom at site \bar{O} . The calculations become simpler if we introduce the variable m_n , defined to be equal to μ_n for Pd atoms and to $\alpha\mu_{Fe}$ for the Fe atom at site \bar{O} . In the absence of an external field, Eq. (2) becomes

$$m_{\bar{n}} = \chi_0 J \sum_{\bar{\delta}} m_{\bar{n}+\bar{\delta}} + \delta_{\bar{n}\bar{O}} (\alpha\mu_{Fe} - \chi_0 JZ_1\mu_1), \quad (5)$$

where μ_1 are the moments of the iron-atom first neighbors. This equation can be solved by a Fourier transformation giving

$$\begin{aligned} m(\vec{K}) &= \sum_{\bar{n}} m_{\bar{n}} e^{i\vec{K}\cdot\bar{n}} \\ &= \alpha\mu_{Fe} - J\chi_0 Z_1\mu_1 + J\chi_0 m(\vec{K}) \sum_{\bar{\delta}} e^{i\vec{K}\cdot\bar{\delta}}, \end{aligned} \quad (6)$$

which can be rewritten as

$$m(\vec{K}) = \frac{\alpha\mu_{Fe} - \Gamma\mu_1}{1 - \Gamma F_1(\vec{K})}, \quad (7)$$

where we have used the first-shell structure factor $F_1(\vec{K}) = Z_1^{-1} \sum_{\bar{\delta}} e^{i\vec{K}\cdot\bar{\delta}}$ and the enhancement parameter Γ . The value of $m_{\bar{n}}$ can be determined by the inverse Fourier transform.

At this point it is convenient to introduce the Fourier coefficients $\Phi_{\bar{n}}$ of the expansion $[1 - \Gamma F_1(\vec{K})]^{-1}$, which are given by

$$\Phi_{\bar{n}} = \frac{1}{V_c^*} \int \frac{e^{-i\vec{K}\cdot\bar{n}} d^3K}{1 - \Gamma F_1(\vec{K})}. \quad (8)$$

The coefficient Φ_0 has an analytic expression for the fcc lattice.⁸ Owing to relation (7), the m_n 's are proportional to the Φ_n 's, therefore

$$m_{\bar{n}} = \frac{m_0}{\Phi_0} \Phi_{\bar{n}} = \frac{\alpha\mu_{Fe}}{\Phi_0} \Phi_{\bar{n}} \quad (9)$$

and

$$m(\vec{K}) = \frac{\alpha\mu_{Fe}}{\Phi_0 [1 - \Gamma F_1(\vec{K})]}. \quad (10)$$

With the scattering of polarized neutrons in single-crystal samples, it is possible to measure the Fourier transforms of the moments⁹

$$M(\vec{K}) = m(\vec{K}) + (1 - \alpha)\mu_{Fe}. \quad (11)$$

The scattering of unpolarized neutrons, on the other hand, is roughly proportional to $[M(\vec{K})]^2$. In each case, for polycrystalline samples, the scattering intensity is proportional to the spherical average of the respective expression, and the large \vec{K} values give μ_{Fe} . Low and Holden³ report a value of $\mu_{Fe} = (3.5 \pm 0.4)\mu_B$. The moment per cloud, $M(0)$, has been determined with neutrons³ or by magnetization measurements^{1,2} as $M(0) = 10\mu_B$. From Eq. (10) we can now see that knowledge of Γ also yields $\alpha\mu_{Fe}$.

The value $\Gamma = 0.947 \pm 0.009$, for pure Pd, was determined by us in a previous work⁷ in which a similar theory was applied to the Ni-Pd alloys. Using these values, we get $\alpha\mu_{Fe} = (0.44 \pm 0.09)\mu_B$. A value of Γ so close to 1 indicates the high polarizability of palladium which is responsible for the giant moments that appear when iron atoms are introduced in this metal. Mueller *et al.*¹⁰ have calculated the band structure of Pd and they have estimated the enhancement factor as $\chi/\chi_0 = 14.9 \pm 1.1$; equating this value to $(1 - \Gamma)^{-1}$, we get $\Gamma = 0.933 \pm 0.005$ which is barely consistent with our value.

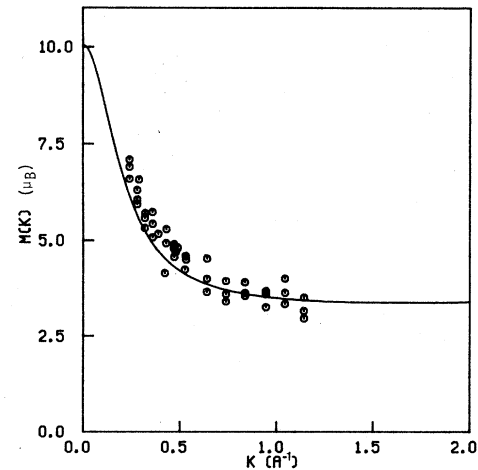


FIG. 1. Spherical average of $M(K)$ using our model (continuous curve) is compared with experimental values (points) from Ref. 4.

We calculated the spherical average of $M(\vec{K})$, using Eq. (11), and compared it with the square root of the experimental values of Hicks *et al.*,⁴ as shown in Fig. 1. The agreement is quite satisfactory. The differences at smaller K could be due to the following: (a) a value of Γ slightly larger than it should be, and (b) the experiment was done with nonpolarized neutrons and therefore some critical scattering could be present, as has been found by Verbeek *et al.*¹¹ in some PdFe alloys.

We also calculated the values for the Φ 's corresponding to our estimated Γ . The calculation was performed by numerical evaluation of the integral of Eq. (8). As shown in Fig. 2, the Φ 's decay exponentially above 2 Å.

III. INTERACTION BETWEEN CLOUDS

The Pd moment-dependent part of the energy for PdFe alloys must be of the following form:

$$E = \sum (1-p_{\vec{n}}) \left[\frac{1}{2} \chi_0 h_{\vec{n}}^2 + \frac{J}{2} \mu_{\vec{n}} \sum_{\vec{\delta}} \mu_{\vec{n}+\vec{\delta}} (1-p_{\vec{n}+\vec{\delta}}) \right] + \dots, \quad (12)$$

where the ellipsis represents higher-order terms in μ . This expression for the energy is consistent with Eq. (1), in fact, Eq. (1) follows from Eq. (12) by minimizing the latter with respect to the moments. Substituting Eqs. (1) and (2) into (12), the following alternative expression for the energy is obtained:

$$E = -\frac{1}{2} \sum (1-p_{\vec{n}}) \mu_{\vec{n}} \left[b_{\vec{n}} + J\alpha \sum_{\vec{\delta}} \mu_{\vec{n}+\vec{\delta}} p_{\vec{n}+\vec{\delta}} \right]. \quad (13)$$

Here we can see that the interaction energy is proportional to the product of the moments of the iron atoms and the first-neighbor palladium atoms. Equations (12) and (13) are valid in the case of a ferromagnetic material where all the moments have the same orientation.

Near the critical temperatures, the orientations of the moments are fluctuating and therefore the model should be modified to take this into account. We then write the iron moment in the following vectorial form:

$$\vec{\mu}_{\vec{m}} = \mu_{\text{Fe}} \vec{\eta}_{\vec{m}}, \quad (14)$$

while the moment of the palladium atoms is as-

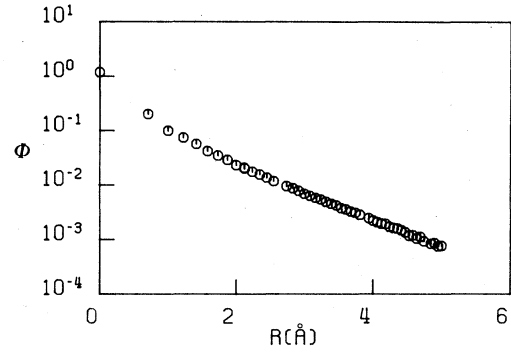


FIG. 2. Calculated values of the Φ 's using our estimated Γ .

sumed to be the vectorial sum of the moments induced by each iron atom:

$$\vec{\mu}_{\vec{m}} = \alpha \mu_{\text{Fe}} \sum_{\vec{l}} p_{\vec{l}} f_{\vec{m}}^{(\vec{l})} \vec{\eta}_{\vec{l}}. \quad (15)$$

Here $f_{\vec{m}}^{(\vec{l})}$ is the magnitude of the moment, in units of $\alpha \mu_{\text{Fe}}$, produced by an iron atom at \vec{l} on a palladium atom at site \vec{m} . The direction of the induced moment is assumed to be, at any time, the same as that of the iron moment.

To obtain a new expression for the energy we substitute the scalar moments, in Eq. (13), by their vector forms, Eqs. (14) and (15), which in the absence of an external field yields

$$E = -\frac{1}{2} \sum_{\vec{l}, \vec{m}} p_{\vec{l}} p_{\vec{m}} J_{\vec{l}, \vec{m}} (\vec{\eta}_{\vec{l}} \cdot \vec{\eta}_{\vec{m}}), \quad (16)$$

where

$$J_{\vec{m}, \vec{l}} = \frac{J_0}{2} \left[\sum_{\vec{\delta}} (1-p_{\vec{m}+\vec{\delta}}) f_{\vec{m}+\vec{\delta}}^{(\vec{l})} + \sum_{\vec{\delta}} (1-p_{\vec{l}+\vec{\delta}}) f_{\vec{l}+\vec{\delta}}^{(\vec{m})} \right] \quad (17)$$

and

$$J_0 = J(\alpha \mu_{\text{Fe}})^2. \quad (18)$$

TABLE I. Values obtained with our model for PdFe alloys.

$\alpha \mu_{\text{Fe}} = (0.44 \pm 0.09) \mu_B$
$\chi_0 = 3.63 \times 10^{-7}$ emu/g
$J = (766 \pm 100)$ K
$J_0 = (147 \pm 34)$ K
$E_c = (-149 \pm 34)$ K
$D/c = (780 \pm 180)$ Å ² K/ at. % Fe

The resulting energy expression indicates a Heisenberg type of interaction between magnetization clouds centered on each iron atom. The total moment associated with each cloud is composed of the central iron moment plus the moments induced by that iron on the palladium atoms.

The $J_{\vec{I}\vec{I}}$ terms correspond to the self-energy of a magnetization cloud. In the case of an isolated iron atom, this energy is given by

$$E_c = -\frac{1}{2}J_0Z_1\frac{\Phi_1}{\Phi_0}. \quad (19)$$

J_0 is a natural unit of energy for the moment dynamics of the system and can be calculated using the value of the susceptibility, $\chi = 6.85 \times 10^{-6}$ emu/g, reported by Verbeek *et al.*¹¹ Using Eqs. (3), (4), (18), and (19), we obtain the values of χ_0, J, J_0 , and E_c given in Table I.

As a first approximation, $f_{\vec{m}}^{(\vec{I})}$ equals $\Phi_{\vec{m}-\vec{I}}/\Phi_0$ when one neglects the presence of iron atoms other than the one located at \vec{I} . In Eq. (17) this is not a good approximation because there is an iron atom as first neighbor of the palladium atom. We can take into account the effect of having an iron atom at site \vec{n} by subtracting the contribution to the moment at \vec{m} due to the palladium substituted by the iron at \vec{n} , that is

$$f_{\vec{m}}^{(\vec{I})} = \Phi_0^{-1}(\Phi_{\vec{m}-\vec{I}} - \Phi_{\vec{n}-\vec{I}}g_{\vec{m}}^{(\vec{n})}), \quad (20)$$

where $g_{\vec{m}}^{(\vec{n})}$ is the normalized moment produced by the palladium atom taking into account the absence

$$J_{\vec{I}\vec{m}} = \frac{J_0}{\Phi_0^2 - \Phi_{\vec{I}-\vec{m}}^2} \sum_{\vec{\delta} \neq \vec{I}-\vec{m}} \Phi_{\vec{m}+\vec{\delta}-\vec{I}}\Phi_0 - \Phi_{\vec{m}-\vec{I}}\Phi_{\vec{\delta}}. \quad (23)$$

This result is valid in the low concentration limit. Note that $J_{\vec{I}\vec{m}}$ is roughly proportional to $\Phi_{\vec{m}-\vec{I}}$. We calculate $J_{\vec{I}\vec{m}}$ using the previously determined Φ 's; the results are shown in Fig. 3, where the continuous curve corresponds to the empirically found fitting function

$$J(R) = J_0 \exp \left[0.406 - 1.105 \frac{R}{a} + \frac{2.508}{(1+R/a)^2} \right], \quad (24)$$

which will be used in the next section.

IV. MONTE CARLO CALCULATIONS

The model developed allows a description of the ferromagnet-superparamagnet transition for a low-

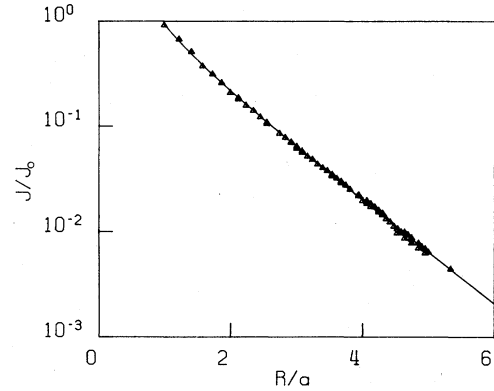


FIG. 3. Values of J calculated using Eq. (23). The continuous curve corresponds to the fitting function given by Eq. (24).

of a palladium at site \vec{I} .

By the above argument we obtain

$$g_{\vec{m}}^{(\vec{n})} = \Phi_0^{-1}(\Phi_{\vec{m}-\vec{n}} - \Phi_{\vec{I}-\vec{n}}f_{\vec{m}}^{(\vec{I})}). \quad (21)$$

Eliminating $g_{\vec{m}}^{(\vec{n})}$ from Eq. (20) we finally have

$$f_{\vec{m}}^{(\vec{I})} = (\Phi_0^2 - \Phi_{\vec{n}-\vec{I}}^2)^{-1} \times (\Phi_{\vec{m}-\vec{I}}\Phi_0 - \Phi_{\vec{n}-\vec{I}}\Phi_{\vec{m}-\vec{n}}). \quad (22)$$

Using this expression, we can write the value of J neglecting all effects due to iron atoms other than the ones at \vec{I} and \vec{m} :

concentration PdFe alloy. From the work of Chouteau and Tournier² we know that above 0.5 at. % Fe the nonlinear interference between magnetization clouds becomes increasingly important as shown by the decrease of the saturation moment per Fe atom; on the other hand, the same authors did not find a ferromagnetic transition below 0.02 at. % Fe, where the alloys are probably spin-glasses.¹² We performed Monte Carlo calculations within this range of concentrations for which the expression for J , Eq. (23), should be valid. When two iron atoms are first neighbors there is an additional interaction, an Fe-Fe exchange, which should be larger than the effective Fe-Fe interaction mediated by the palladium atoms. Given the low occurrence of iron pairs at this concentration, we completely neglected this effect.

A description of the Monte Carlo method can be found in the book by Binder.¹³ N classical spins of magnitude 1 were distributed at random in a cube of a size determined by the concentration. The use of classical spins in the calculations should be a good approximation at high temperatures because of the large magnetic moments of the clouds ($\sim 10\mu_B$) which correspond, for $g=2$, to a spin of $S=5$. Given the small concentrations we neglected the crystal lattice. The exchange constants $J(R)$ were calculated using Eq. (24), and the energy using Eq. (16) (excluding the formation energy of magnetization clouds). Periodic boundary conditions were used.

For each concentration, we computed for various temperatures: the energy E , a component M_z of the total magnetic moment, the average magnitude (M) of the total magnetic moment (\bar{M}), the specific heat (from the average energy fluctuation), the susceptibility χ , (from the average fluctuation of a component of the total magnetic moment), and a quantity $\tilde{\chi}$ which coincides with χ for infinite systems and which is given by the fluctuation of the magnitude of the total moment, that is,

$$\tilde{\chi} = \frac{\langle (M - \langle M \rangle)^2 \rangle}{NkT}. \quad (25)$$

The calculations were performed in the following way:

- (1) For most concentrations, $N=50$ was used.
- (2) For each temperature, and in order to ensure the attainment of equilibrium, 500 Monte Carlo steps per spin were performed before calculating the averages.
- (3) At each Monte Carlo step per spin the instantaneous values of the energy and the total magnetic moment were recorded. 1500 of those values were used to compute the averages.
- (4) The initial configuration of spins, for each temperature, was set equal to the final configuration of the previous temperature. We found the same results lowering or raising the temperature.
- (5) For concentrations $c=0.0592$, 0.0430, and 0.0287 at. % Fe, systems of $N=100$ spins and 4000 instantaneous values were used.

For systems as small as these, and for any temperature, the total magnetic moment rotates so that any component of the moment averages to zero after a certain elapsed time. This makes very difficult the estimation of the critical temperatures from the "true" susceptibility χ . On the other hand, the average value of the magnitude of the magnetization changes too smoothly with temperature to al-

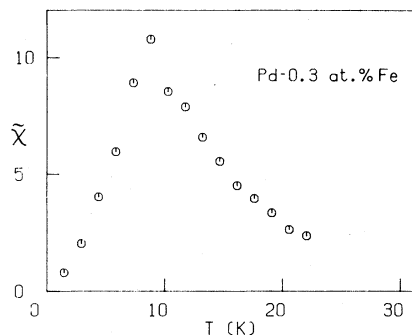


FIG. 4. Typical $\tilde{\chi}$ vs T curve. Critical temperatures were determined from the maximum of $\tilde{\chi}$.

low a precise determination of the critical temperatures. Nor can we determine these temperatures from the specific heats calculated as fluctuations of the energy because the large dispersion of the values disguises the peak at the critical temperature. The critical temperatures were therefore determined from the maximum of $\tilde{\chi}$. A typical $\tilde{\chi}$ vs T curve is shown in Fig. 4. At the lowest concentration it is difficult to determine the positions of the maxima of $\tilde{\chi}$ vs T , consequently we also used the specific heat determined by the derivation of a cubic spline¹⁴ interpolation of the energy; this was done for $c=0.0287$, 0.0430, and 0.0592 at. % Fe. The $\tilde{\chi}$ vs T curve for the latter concentration is shown in Fig. 5 while the specific heat and the energies are shown in Fig. 6. The calculated susceptibilities and specific heat are adequate to determine the critical temperatures but, because of the use of *classical* spins, they are not a good representation of the behavior of the real systems at lower temperatures.

The critical temperatures obtained with this procedure are shown in Fig. 7 together with the experimental values.^{1,2,15-19} There is very good agree-

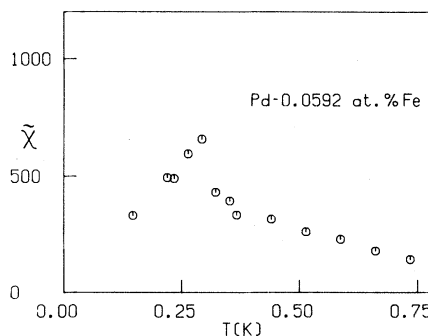


FIG. 5. $\tilde{\chi}$ vs T curve for $c=0.0592$ at. % Fe. Below this concentration, the critical temperatures were difficult to determine using the maximum of $\tilde{\chi}$.

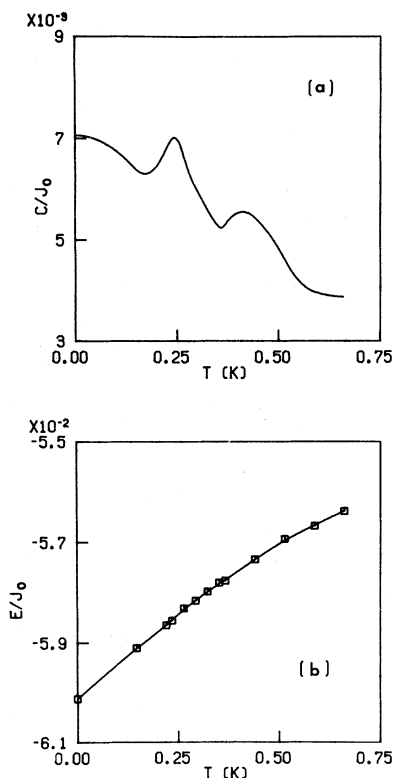


FIG. 6. (a) Specific-heat and (b) energy curves for $c=0.0592$ at. % Fe. The maximum of C/J_0 was also used to determine the critical temperature of this alloy.

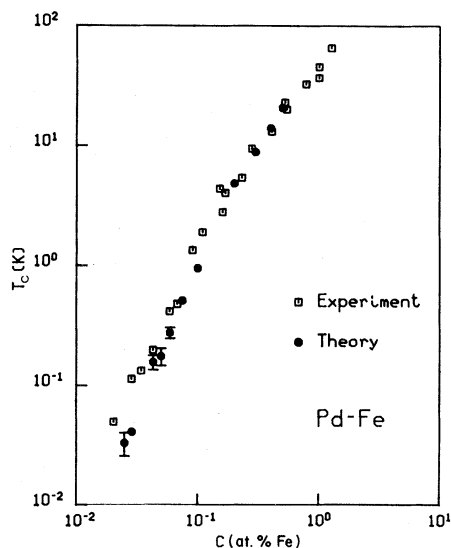


FIG. 7. Critical temperatures determined with our model by Monte Carlo calculations are compared with experimental results from Refs. 1, 2, and 13–19.

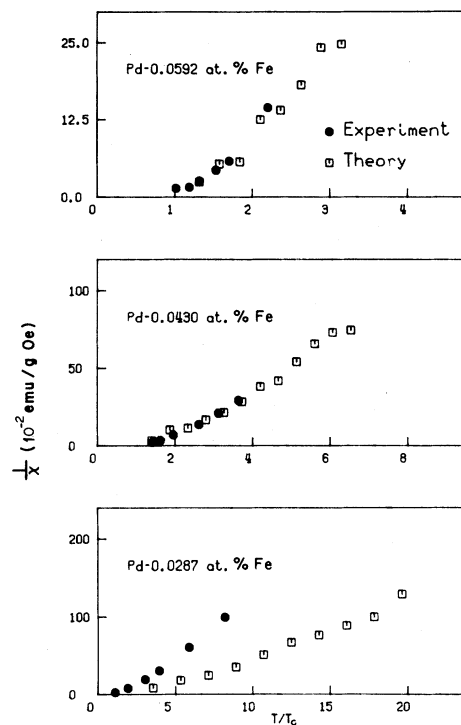


FIG. 8. Comparison of calculated magnetic susceptibilities with the experimental values from Ref. 2.

ment for a wide range of concentrations; only for $c=0.0287$ and $c=0.025$ at. % Fe the calculated T_c 's are smaller than the experimental ones. The experimental T_c is proportional to c^2 for $c < 0.1$ at. % Fe, while above this concentration, the T_c vs c curve is well approximated by a straight line. All these features are also shown by our calculated values. In our model the shape of the curve T_c vs c is essentially determined by the spatial extent of the magnetization clouds, i.e., by the value of Γ . Not only is the shape of the curve reproduced by the calculations but also the values of T_c which are proportional to J . This result checks our value of J which was determined independently.

We also compared some calculated susceptibilities above T_c with experimental values, as shown in Fig. 8. The susceptibility per cloud was calculated using the following expression:

$$\chi = \frac{S+1}{S} \frac{M(0)^2 \langle M_z^2 \rangle}{NkT}, \quad (26)$$

where the quantum nature of the spin was taken into account by the factor $(S+1)/S$. The temperatures were scaled with the critical temperatures. Although the experimental and calculated values of the critical temperatures are close, they are not

identical and therefore this procedure makes the comparison easier. There is very good agreement for the two larger concentrations. As in the case of the critical temperatures, at $c=0.0287$ at. % Fe there is no agreement with experimental values.

V. OTHER RESULTS

The same model can be used for concentrated Pd-Fe alloys if we include an environment-dependent iron moment. For example, we may apply the model to the ordered alloy FePd₃. From Eqs. (1), (2), and (4) one obtains in this case

$$\mu_{\text{Pd}} = \frac{\Gamma \alpha \mu_{\text{Fe}}}{3 - 2\Gamma} \quad (27)$$

For this alloy, Shirane *et al.*²⁰ measured $\mu_{\text{Fe}} = (2.86 \pm 0.01) \mu_B$. Using this value of the iron moment, we obtain $\mu_{\text{Pd}} = (0.31 \pm 0.08) \mu_B$, while Shirane *et al.* give the value $\mu_{\text{Pd}} = (0.34 \pm 0.01) \mu_B$.

Another quantity that can be estimated with our model is the stiffness coefficient D of acoustics spin waves for low-concentration PdFe alloys. From the Hamiltonian of Eq. (16), we obtain the stiffness coefficient from the expression

$$D = \frac{c}{6(S+1)} \sum_{\vec{n}} J_{\vec{0}\vec{n}} |\vec{n}|^2 \quad (28)$$

This is the usual expression for a Heisenberg Hamiltonian,²¹ except that the random position of the iron atoms, in the lowest order, gives rise to the factor c .

We calculated D using the J 's of Eq. (23) and neglecting the unknown Fe-Fe first-neighbor exchange. We obtain $D/c = (780 \pm 180) \text{ \AA}^2 \text{ K/at. \% Fe}$. This value is consistent with the one estimated by Smith *et al.*²² [$D/c = (690 \pm 35) \text{ \AA}^2 \text{ K/at. \% Fe}$], who included in the calculation their own heat-capacity data and other experimental results, such as the neutron scattering measurements of Stringfellow.²¹

VI. CONCLUSIONS

The model we have presented describes well several magnetic properties such as critical temperatures, shape of the magnetization cloud, magnetic susceptibilities, and spin-wave stiffness constant. This model agrees with the measurements of Chouteau and Tournier² in that this system is a ferromagnet for $c > 0.02$ at. % Fe. Therefore, although there is a change in the behavior of T_c near $c = 0.1$ at. % Fe, this is not a true critical concentration. Our model fails for concentrations below 0.03 at. % Fe. This is probably due to a competition between the ferromagnetic and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions which are of longer range. Below 0.02 at. % Fe, RKKY interactions probably dominate making the system a spin-glass.¹²

The success of our model supports the idea of polarization clouds produced by first-neighbor interactions between localized moments. This is not in contradiction with the idea of a nonlocal susceptibility since the latter is proportional to our $\Phi_{\vec{n}}$. However, we may conclude that the high degree of localization of d electrons in Pd should be taken into account for a proper calculation of the nonlocal susceptibility from first principles. We do not believe, therefore, that the magnetization cloud could be due to an enhancement of RKKY interactions as suggested by Verbeek *et al.*¹¹

As a final comment, we may say that a first-principles theory now only needs to justify the local character of the Pd moment in order to agree with a wide range of experimental data.

ACKNOWLEDGMENT

We would like to thank Dr. J. F. Fernández for helpful comments and for his critical reading of this paper.

¹J. Crangle and W. R. Scott, J. Appl. Phys. **36**, 921 (1965).

²G. Chouteau and R. Tournier, J. Phys. (Paris), Colloq. **32**, C1-1002 (1971).

³G. G. Low and T. M. Holden, Proc. Phys. Soc. London **89**, 119 (1966).

⁴T. J. Hicks, T. M. Holden, and G. G. Low, J. Phys. C **1**, 528 (1968).

⁵S. Doniach and E. P. Wohlfarth, Proc. R. Soc. London

Ser. A **296**, 442 (1967).

⁶M. J. Zuckermann, Solid State Commun. **9**, 1861 (1971).

⁷R. E. Parra and R. Medina, Phys. Rev. B **22**, 5460 (1980).

⁸G. S. Joyce, J. Phys. C **4**, 153 (1971).

⁹R. Medina and J. W. Cable, Phys. Rev. B **15**, 1539 (1977).

¹⁰F. Mueller, A. Freeman, J. Dimmock, and A. Furdyna,

- Phys. Rev. B 1, 4617 (1970).
- ¹¹B. H. Verbeek, G. J. Nieuwenhuys, J. A. Mydosh, C. van Dijk, and B. D. Rainford, Phys. Rev. B 22, 5426 (1980).
- ¹²K. Nagamine, N. Nishida, S. Nagamiya, O. Hashimoto, and T. Yamazaki, Phys. Rev. Lett. 38, 99 (1977).
- ¹³K. Binder, in *Monte Carlo Methods in Statistical Physics*, edited by K. Binder (Springer, Berlin, 1979), p. 1.
- ¹⁴C. H. Reinsch, Numer. Math. 16, 451 (1971).
- ¹⁵J. Crangle, Philos. Mag. 5, 335 (1960).
- ¹⁶G. Williams and J. W. Loram, J. Phys. Chem. Solids 30, 1827 (1969).
- ¹⁷P. E. Clark and R. E. Meads, J. Phys. C 3, S310 (1970).
- ¹⁸G. Longworth and C. C. Tsuei, Phys. Lett. 27A, 258 (1968).
- ¹⁹A. J. Manuel and M. McDougald, J. Phys. C 3, 147 (1970).
- ²⁰G. Shirane, R. Nathans, S. J. Pickart, and H. A. Alperin, in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (IOP, London, 1965), p. 223.
- ²¹M. W. Stringfellow, J. Phys. C 1, 1699 (1968).
- ²²T. F. Smith, W. E. Gardner, and H. Montgomery, J. Phys. C 3, S370 (1970).