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Electrical Resistivity of *Pd*Fe Alloys*†

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The electrical resistivity ρ of a series of ferromagnetic *Pd*Fe alloys varying in composition from 1 to 12 at.% Fe was measured from 4.2 to 300 °K. The data were analyzed by subtracting from the measured ρ values, the value of the electrical resistivity of a sample of 99.999+%-purity Pd at each temperature. From these values of the incremental resistivity $\Delta\rho$ we have obtained the temperature-dependent part by subtracting its limiting low temperature value (at 4.2-°K). The reuslting temperature- and concentration-dependent resistivity ρ_m has the following properties: For alloys of Fe concentration $c \gtrsim 2$ at.%, $\rho_m \propto AT^2$ up to about 40 °K in all but the highest-concentration alloys studied. Above this temperature we find a $T^{3/2}$ law up to the ferromagnetic ordering temperature T_C except in alloys with $c \gtrsim 6$ at.%, where the temperature dependence is faster above $\approx 0.7T_C$. A 1 at.% alloy sample had a low-temperature behavior intermediate between the T^2 behavior of the higher-concentration alloys and the $T^{3/2}$ behavior observed in alloys with Fe concentration less than 1 at.%. The coefficient A of the low-temperature T^2 dependence of ρ_m increases approximately linearly with Fe concentration, and at 2 at.% is about seven times as large as the value found in pure Pd. These results suggest that electron-magnon scattering is the dominant scattering mechanism determining ρ_m. Our data, above 2 at.% Fe, are best interpreted in terms of a model in which the *Pd*Fe alloys are considered to be approximately magnetically homogeneous with an effective, concentration-dependent "*s-d*" exchange interaction. In terms of this model, the expected correlation between the coefficient A and the spin-wave excitation spectrum, at low temperatures, is shown to exist.

I. INTRODUCTION

There has been considerable interest in the characteristics of magnetic alloys ranging from systems in which a long-range magnetically ordered state is observed to systems in which a spin-compensated state about the magnetic impurity is believed to occur. Of special interest is the electron scattering as measured by the transport coefficients in these magnetic systems. Much effort has been devoted to the study of transport properties in alloys with exchange enhanced matrices such as Pd and Pt. In the low-concentration range, Ni in Pd shows no long-range order for less than 2 at.% and the behavior of this system presently has been described in terms of paramagnons. On the other hand, solutions of Fe in Pd exhibit a long-range ferromagnetic behavior down to very low concentrations. In this paper we describe the results of a systematic study of the electrical resistivity ρ of a series of *Pd*Fe alloys.

The ferromangetic ordering temperature of *Pd* Fe alloys is a very rapidly increasing function of iron concentration. An interesting feature of this system is the interplay of a local moment (Fe) and a nearly ferromagnetic matrix (Pd). At very low iron concentrations the matrix has a very nonuniform spin polarization which becomes essentially uniform at an iron concentration of 3-4 at.%.[1] The giant moments[2] observed in this system at low Fe concentrations result from the polarization of the Pd atoms neighboring an Fe impurity. Since the range of the polarization is quite large, the number of Pd atoms participating in the polarization is also large. The long range of the matrix polarization is due to an exchange enhancement of the Pd susceptibility. The ferromagnetic ordering for low Fe concentrations has been described in terms of a model of interacting clusters of spins centered about the impurity sites.[2,3] However, this cluster model is clearly inappropriate once the matrix polarization becomes essentially uniform.

As the iron concentration is increased to about 8 at.%, each Fe or Pd atom has on the average, in a random alloy, one Fe near neighbor. Certainly when this concentration is reached, one must worry about impurity-impurity interaction effects. Furthermore, at about 10-12 at.% the matrix polarization is essentially saturated at a value of about 0.4 μ_B per Pd atom.[2] It is clear, therefore, that no single model could describe this complex system over the entire concentration range up to and above 12 at.% Fe. In fact, a model which distinquishes the different concentration regions, seems to be most appropriate for this system.

Previously, we have reported for *Pd*Fe the concentration dependence of the magnetic ordering temperature T_C[4] and the critical behavior of $d\rho/dT$ in the neighborhood of T_C.[5] In this paper we present our experimental results for the temperature and concentration dependence of ρ at low temperatures (below the critical region). The range of Fe concentrations reported here extends from 1 to 12 at.%. During the course of our work a similar study of *Pd*Fe alloys for concentrations below 1 at. % was reported by Williams and Loram.[6] In contrast to their observed $T^{3/2}$ behavior of the magnetic component of ρ for concentrations below 1 at. %, we find a low-temperature limiting behavior T^2 for alloys of Fe concentration 2 at. % and greater. The concentration range suudied here is probably outside what one would consider to be the dilute impurity limit so that it is not unexpected that there be some differences between these results and those on the low-concentration alloys. The fact that the magnetic ordering temperature varies linearly with concentration above 1 at. % (with a break in slope at about 4%) suggests that it would be fruitful to study the detailed behavior of ρ over the whole concentration range above 1 at. %. Our main purpose then is to determine the behavior of ρ in a concentration range where the matrix polarization gradually loses its low-concentration inhomogeneity and approaches the situation of a saturated uniform matrix polarization with interacting local moments. In Sec. IV we discuss some conclusions that can be drawn about this system on the basis of these and other experimental results and the present theoretical understanding of the problem.

II. EXPERIMENTAL PROCEDURE

The electrical resistivity was measured as a function of temperature for a series of nine *Pd*Fe alloys varying in composition from 1 to 12 at. % Fe. The measurements were made over a temperature range from 4. 2 to 300 °K except for the 12% sample where the measurements extended to 350 °K. Samples used in this study were usually about 3×10^{-3} cm thick, 2 mm wide, and 2 cm long and were prepared from homogenized ingots. The composition of the samples was taken to be the same as the composition of the starting materials from which the ingots were prepared. A standard four-point probe technique was used to measure the resistivity. The dc current was maintained constant to about one part in 50 000 and the voltage was measured with a precision of a few nanovolts. Between 4. 2 and 100 °K, a Ge resistance thermometer was used to measure the temperature and from 77 °K upwards a copper-constantan thermocouple was used. For each reading at a particular temperature, the temperature was maintained constant to better than 0. 01 °K. From 4. 2 to 20 °K and in the vicinity of the magnetic ordering transition, data were generally taken at 1 °K intervals. In the remainder of the temperature range studied, an interval of 2 or 3 °K was generally used except for above the magnetic ordering temperature where a 5 °K interval was used.

III. EXPERIMENTAL RESULTS

Our results for the electrical resistivity ρ for five of our samples are shown in Fig. 1. There is, in each of the curves, a break in ρ in the vicinity of the ferromagnetic transition temperature T_C which is indicated in the figure by an arrow. T_C has been determined to be the temperature of the maximum in $d\rho/dT$.[4] At temperatures well above T_C, the electrical resistivity depends linearly on temperature as one would normally expect for high-temperature electron-phonon scattering.

We assume that the electrical resistivity ρ (T, c) for an alloy of iron concentration c can be written as

$$\rho(T,c)=\rho_0(T)+\Delta\rho, \tag{1}$$

where $\rho_0(T)$ is the pure Pd resistivity, and $\Delta\rho$ is the temperature- and concentration-dependent contribution to the resistivity which is due to the presence of the magnetic impurity Fe. In $\Delta\rho$ there is a contribution from potential scattering which is temperature independent; the remainder might be attributed entirely to spin-dependent scattering mechanisms. This will be strictly true provided that the presence of Fe impurities in the Pd matrix does not change significantly any other mechanisms, e.g., electron-electron or electron-phonon scat-

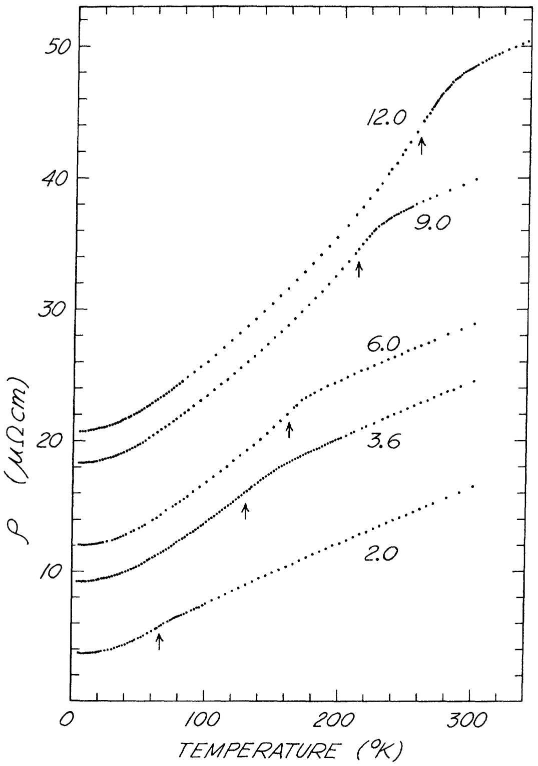

FIG. 1. Electrical resistivity ρ of a series of *Pd*Fe alloys as a function of temperature. The arrows locate the respective transition temperatures. The numbers associated with each curve are the Fe concentrations in at. %.

tering which contribute to the total resistivity of this system.

Our main interest is in the way that the long- or short-range spin order in the *Pd*Fe system affects the electrical resistivity. We believe that the dominant contribution to the temperature-dependent part of $\Delta\rho$ arises from this spin disordering and we have carried out the analysis of the data accordingly.

We have therefore measured the resistivity of a sample of Pd of 99.999+%-purity and have used these values to substitute in Eq. (1) for $\rho_0(T)$. Thus to get the total impurity-dependent resistivity $\Delta\rho$, we have subtracted from the measured resistivity the measured pure Pd resistivity at each temperature. Since we are not interested in the potential scattering part of $\Delta\rho$ we subtract from $\Delta\rho$ at each temperature its value at 4.2 °K, the lowest temperature used in these experiments. In Fig. 2 we show the temperature dependence of $\rho_m \equiv \Delta\rho \times (T, c) - \Delta\rho(4.2\,^\circ\mathrm{K},\ c)$ for several values of the Fe concentration.

The curves in Fig. 2 show a pronounced change in slope at the ferromagnetic ordering temperature T_C which would correspond to the temperature at which the slope of these curves has its maximum value. Far above T_C, the curves are essentially flat, we have found no evidence of a temperature- or concentration-dependent scattering mechanism above about $T \approx 2T_C$ which cannot be accounted for by the temperature dependence of the pure-Pd resistivity $\rho_0(T)$ in this temperature range. In the interval between T_C and the flat portion of the curves, there is a temperature-dependent contribution to ρ_m which arises from the short-range magnetic order that extends above T_C.

Below T_C the temperature dependence of ρ_m is complex and does not follow a simple power law over the entire temperature range. By using log-log plots of ρ_m versus T/T_C we can fit the data to $\rho_m \propto T^2$ up to approximately 40 °K in all samples other than the 1 and 12 at. % samples, and to $\rho_m \propto T^{3/2}$ above this temperature up to the ordering temperature in all but the highest-concentration samples (7.5, 9, and 12 at. %). For these latter samples the resistivity varies faster than $T^{3/2}$ (in fact, more like T^2) in the interval from about $0.7T_c$ to T_c. The 1 at. % sample does not show the T^2 low-temperature behavior but is intermediate between $T^{3/2}$ and T^2.

The low-temperature (below ≈50 °K) behavior of ρ_m is shown in Fig. 3 by plotting ρ_m versus T^2 for several of the samples studied. It is clear from this figure that the fit to a T^2 dependence is excellent and extends over a very substantial temperature interval. The departure from a T^2 dependence occurs at about 40 °K in all but the highest-concentration samples, a very striking and unexpected effect which is probably associated with a property of the Pd matrix as influenced by the magnetic impurities.

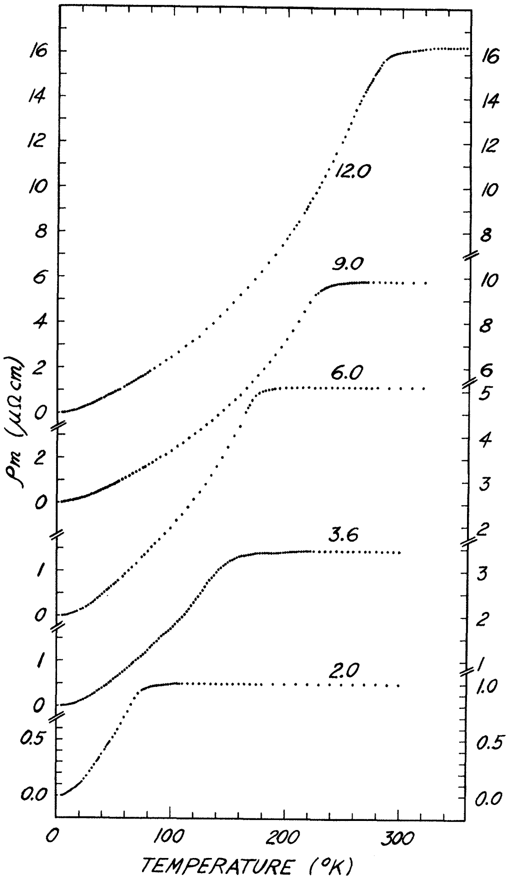

FIG. 2. ρ_m as defined in the text, for the *Pd*Fe alloys shown in Fig. 1, as a function of temperature. The numbers associated with each curve are the Fe concentrations in at.%. Note the scale changes for the different concentrations.

The concentration dependence and magnitude of the coefficient A of the low-temperature T^2 dependence of ρ_m should shed valuable light on the nature of the scattering mechanism(s) present. The concentration dependence of A obtained from the data in Fig. 3 is shown in Fig. 4. The solid curve shown is the best straight-line fit to the data points with little weight given to the $7\frac{1}{2}$% point because of the relatively large uncertainty in the dimensions of this particular sample. As is evident from the Fig. 4, A increases approximately linearly with concentration over the entire range of concentrations studied here. As mentioned previously, the 1% sample does not appear here since the observed low-temperature behavior was considerably different from T^2. The magnitude of A for our lowest-concentration sample (2 at. %) is about a factor of 7 larger than the value observed in pure Pd (0.31×10^{-6} $\mu\,\Omega$cm/°K^2 for T

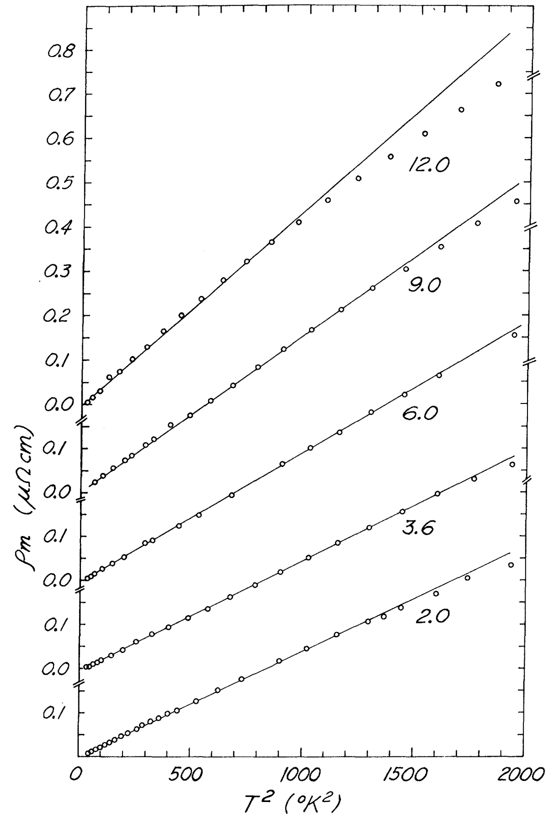

FIG. 3. ρ_m versus T^2, otherwise as in Fig. 2.

less than 10 °K).[7]

Another characteristic of the ρ_m behavior which should be accessible to theoretical prediction is the absolute change in ρ_m between T_C and 0 °K. In Fig. 5 we show our results for $\Delta\rho_m \equiv \rho_m(T_c, c) - \rho_m(4.2\ °\mathrm{K}, c)$ as a function of the square of the Fe concentration. Note that $\Delta\rho_m$ is calculated using for T_C the value of T for which the slope in ρ_m is maximum. The solid lines in Fig. 5 are shown to indicate the trend of the data. There is a sharp kink in the data points occurring between 3 and 4 at.%. It is about this concentration that there is also a change in slope of the linear dependence of T_C on concentration. Our value for $\Delta\rho_m$ for the 1% sample is consistent with the value to be expected from the trend of $\Delta\rho_m$ observed in Ref. 6 at lower concentration.

IV. DISCUSSION

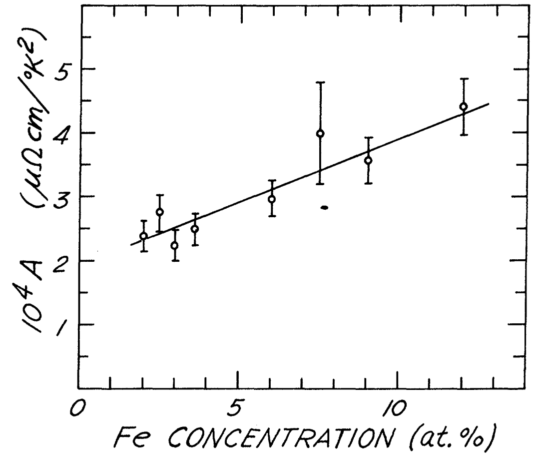

FIG. 4. The coefficient of the low-temperature T^2 dependence of ρ_m as a function of Fe concentration. The solid curve is the best straight-line fit to the data.

The complexity of the observed temperature and concentration dependence of the electrical resistivity of the alloys of *Pd*Fe studied here would seem to preclude a simple comprehensive explanation of all of our data. In particular, the temperature dependence of ρ_m indicates that a change in the dominant scattering mechanisms occurs at a temperature of approximately 40 °K in almost all of our alloys. Furthermore, the temperature dependence of ρ_m for the highest-concentration alloys (above 6%) appears to be different from that of the lower-concentration alloys for $0.7T_c \lesssim T \lesssim T_C$, whereas our previous measurements[5] in the critical region immediately *above* T_C indicated a very *similar* behavior in alloys containing 3, 6, and 9 at.% Fe. The

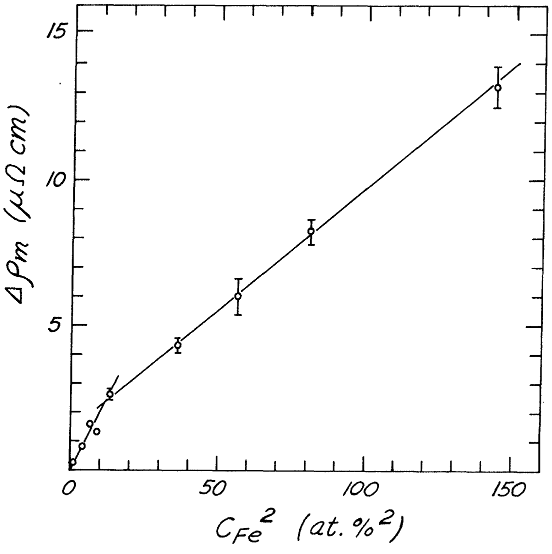

FIG. 5. $\Delta\rho_m \equiv \rho_m(T_C) - \rho_m(4.2\ °\mathrm{K})$ as a function of the square of the Fe concentration. The slopes of the solid lines are 0.21 $\mu\Omega$ cm/(at.%)^2 and 0.083 $\mu\Omega$ cm/(at.%)^2.

absence of any complete theoretical understanding of the relevant scattering mechanisms (even in a homogeneous elemental ferromagnet) when T/T_C begins to be comparable to unity, makes it difficult to specify fully the reasons for the observed behavior. The following remarks can be made, however, about the entire temperature interval from about 40 °K to T_C for all of our alloys with Fe concentrations greater than 1 at.%, (with the possible exception of the highest concentrations $\gtrsim 9$ at.%).

First of all, the lower bound for this temperature interval seems to be determined by some property of the matrix since an abrupt change in ρ_m occurs at this temperature in all alloys. Recently a prediction of the temperature dependence of the electronic contribution to the specific heat C_e of pure Pd has been obtained from the measured heat capacity and calculated lattice contribution using the results of neutron-scattering experiments.[8] The temperature dependence of C_e shows considerable structure, and in particular above 20 °K deviates from the low-temperature-limiting value of γT obtained by Veal and Rayne.[9] In fact, one has effectively a lower value of γ by about 30% somewhat above this temperature. If electron-electron scattering as proposed by Rice[10] makes a significant contribution to the electrical resistivity of the transition metals, and if, as he suggests, the coefficient of T^2 depends on the square of the electronic specific-heat coefficient γ, then the sharp change in the ρ_m behavior of our alloys in this temperature region (≈ 40 °K) may be connected with a change in γ. On the other hand, our values for the low-temperature coefficient of the T^2 behavior of ρ_m are almost an order of magnitude larger than in pure Pd and furthermore this coefficient increases with increasing Fe concentration, whereas the observed γ decreases with increasing Fe concentration.[9] Thus we must conclude that the observed T^2 behavior in our alloy is probably not predominantly an electron-electron scattering effect. However, the possibility exists that there is some electron-electron scattering present which may affect the region over which a T^2 dependence of ρ_m is observed in the alloys. We shall come back to this point a bit later.

Second, a linear dependence of $\Delta\rho$ on temperature is not observed immediately below T_C, in contrast to the situation in very dilute alloys[6] (less than 1 at.%) where such a linear dependence was observed and is theoretically predicted. We shall not comment further about the higher-temperature behavior of ρ_m but will try to draw some conclusions about *Pd*Fe alloys from the observed low-temperature T^2 behavior.

A number of mechanisms have been proposed to explain the observed T^2 low-temperature behavior of the electrical resistivity of transition elements. Among these are electron-electron scattering[10-12] and spin-wave–induced electron scattering.[13-15] In addition, electron-paramagnon scattering has been advanced as a possible T^2 mechanism in paramagnetic transition-metal alloys.[16,17] As we have already indicated, the magnitude and concentration dependence of the coefficient A of the T^2 dependence of ρ_m appears to be much different from that to be expected from purely electron-electron scattering. We believe that the observed low-temperature behavior of ρ_m in our ferromagnetically ordered alloys is due to spin-wave–induced electron scattering (electron-magnon scattering).

It has been predicted recently[18] that, in alloys, electron-magnon scattering leads to a $T^{3/2}$ low-temperature ($T/T_C \ll 1$) dependence as opposed to a T^2 dependence of ρ_m on T because of the lack of translational invariance in alloy systems. Indeed, such a temperature dependence was observed in *Pd* Fe for $c < 1$ at.% as reported in Ref. 6. Our results which give a limiting T^2 behavior for ρ_m above 1 at.%. thus indicate that there is a clear distinction between the very dilute alloys and the higher concentration alloys of *Pd*Fe. Our 1 at.% data for ρ_m show that this concentration alloy, which has a limiting $T^{1.75}$ behavior, straddles the two concentration regions. As we have indicated in the Introduction, it is not surprising that the very dilute alloys of *Pd*Fe behave quite differently from the higher-concentration alloys since the very dilute alloys do possess a very inhomogeneous matrix spin polarization which is gradually smoothed out as the Fe concentration increases. Several experimentally determined properties, the magnetic ordering temperature,[4] the spin-wave stiffness constant D,[19] and the high-field susceptibility χ_{HF} show[20] a marked change in their concentration dependence at about 1–2 at.%.

Our result that the limiting low-temperature behavior of ρ_m follows a T^2 law above 1 at.% as opposed to the observed $T^{3/2}$ law below 1 at.%, suggests that the lack of translational invariance in the *Pd*Fe alloy system does not fundamentally alter the characteristic temperature dependence of the electron-magnon scattering from that predicted for homogeneous ferromagnets. We believe that the reason for this is that in *Pd*Fe alloys, where there is a strong matrix polarization mechanism, there is necessarily a considerable degree of magnetic homogeneity, certainly above 2 at.% or so. The best evidence of this magnetic homogeneity, besides the measurements reported here, is the fact that the neutron scattering experiments[1] display considerable homogeneity for the matrix polarization as one goes above 1–2 at.% Fe, and the linear dependence on concentration of the magnetic-ordering temperature above 1 at.%.[4] We feel confident that in this concentration range, *Pd*Fe alloys have many of the characteristics of a nearly homogeneous ferromagnet rather than being describable as a very inhomogeneous collection of giant moment clusters in a

Pd matrix. This is not to say that purely local effects, such as the direct exchange interaction between an Fe atom and its nearest-neighbor Pd atoms, do not play a role. It does appear surprising, however, that the T^2 dependence persists to such high reduced temperatures ($T/T_C \approx 0.5-0.6$) in the lowest-concentration alloys studied since one would expect that spin-wave excitations would not be very well defined for such high reduced temperatures. We have no explanation for this apparent discrepancy.

If, as we argue above, one is to consider the *Pd*Fe alloys above ≈2 at.% as nearly homogeneous ferromagnets, we should be able to compare our results for the coefficients A of the low-temperature T^2 dependence of ρ_m with the theory of electron-magnon scattering in homogeneous ferromagnets as given, for example, in Ref. 15. We must consider, however, that the important physical parameters such as the s-d exchange integral J and the spin-wave stiffness constant D are probably concentration dependent. One has from Ref. 15 that A should be proportional to J^2/D^2. Since D is an increasing function of Fe concentration in this concentration range,[19] it is clear that we cannot obtain agreement with our observed behavior of A unless J^2 were considered to be an increasing function of concentration also. Values of J were deduced from the dilute-alloy studies of Ref. 6 and J was shown to increase only by about 20% from 0.16 to 0.78 at.%. A change of this order of magnitude is insufficient to account for the change in A with concentration observed by us for higher concentrations. However, this estimate of the concentration dependence of J based on measurements below 1 at.% does not conclusively eliminate the possibility of a more rapid dependence on J on concentration above 1 at.%. Thus, whether one can consider J to be an effective concentration-dependent s-d exchange integral is still an open question which will probably only be resolved when a more detailed treatment of the electron-magnon scattering for systems like *Pd*Fe is available.

We do have an additional set of data, namely, the concentration dependence of $\Delta\rho_m$ which is a measure of the total spin-disorder scattering if no other scattering mechanisms are present to a significant degree. Yosida[21] and Kasuya[22] have predicted that for a dilute alloy, $\Delta\rho_m$ is proportional to J^2c. However, this relationship is probably not applicable to the concentration range studied here, even if it applied at all in a highly polarizable system like *Pd*Fe. To be consistent with the suggestion that the alloys studied here approximate homogeneous ferromagnets, we can again use the theoretical prediction for homogeneous ferromagnets[23] that $\Delta\rho_m$ is proportional to J^2, and interpret J as a concentration-dependent effective s-d exchange integral. Eliminating J^2 between the two relationships involving A and $\Delta\rho_m$, we obtain

$$\Delta\rho_m/A \propto D^2. \tag{2}$$

The values of $\Delta\rho_m/A$ calculated from our data are shown as a function of concentration in Fig. 6. Although it is not obvious from the dependence of $\Delta\rho_m$ and A on concentration shown in Figs. 4 and 5, the ratio $\Delta\rho_m/A$ has a very simple linear dependence on concentration over the entire range from 2 to 12 at.%. (If A and $\Delta\rho_m$ varied precisely as shown by the straight lines in Figs. 4 and 5, respectively, the calculated values of $\Delta\rho_m/A$ would be within a few percent of the straight line shown in Fig. 6 over the range of concentrations from 2.5 to 12 at.%.) The only available data for D in this concentration range (2.2, 4.0, and 10.0 at.%) due to Stringfellow[19] is consistent with a linear increase of D^2 with c above 2 at.%.

We conclude that there is therefore some justification for our point of view that the gross features of the temperature and concentration dependence of the electrical resistivity of *Pd*Fe alloys in the concentration range above 2 at.% are consistent with an interpretation of these alloys as nearly homogeneous ferromagnets. Furthermore, the predominant scattering mechanism contributing to ρ_m is spin-disorder scattering, i.e., s-electron scattering from spin waves of the coupled Fe spin-Pd d spin system. A detailed comparison of our experiments with theory is not possible since

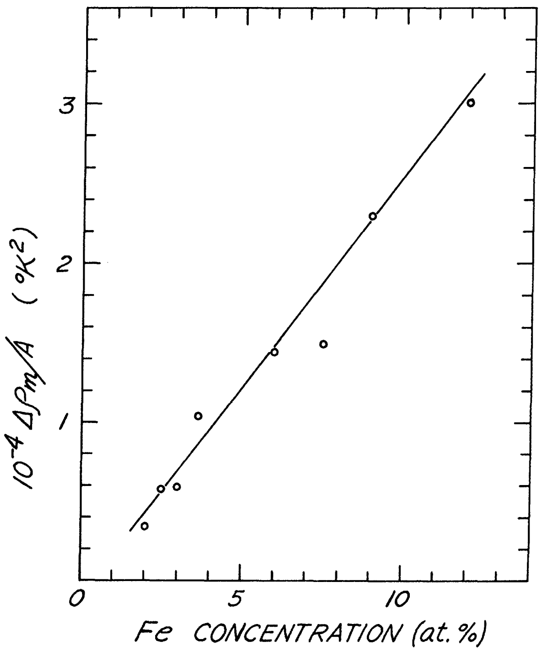

FIG. 6. $\Delta\rho_m/A$ calculated from the data shown in Figs. 4 and 5 as a function of Fe concentration. The solid curve is the best straight-line fit to the data.

a theory does not exist for the type of system studied here. However, the apparently simple relation between the coefficient of the T^2 low-temperature dependence of ρ_m, the total spin-disorder scattering $\Delta\rho_m$ and the iron concentration, together with the consistency of the interpretation of our data in terms of electron-magnon scattering with an effective concentration dependent *s-d* exchange integral, suggest that such a theory should be obtainable from a complete treatment of the *s-d* scattering mechanisms in terms of the dynamic susceptibility $\chi(\vec{q}, \omega)$ for *Pd*Fe. A recent paper[24] has considered in some detail the properties of this function and shows that the spin-wave excitations are damped. This should have an important effect on the electron-magnon scattering.

Note added in proof. All of the experiments reported in this paper were performed in zero applied magnetic field. The effect of the demagnetizing field on the resistance is not considered in this study.

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Band-Structure Effects in Itinerant Antiferromagnetism

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The effect of imperfect "nesting" of the band structure on itinerant antiferromagnetism is considered. A model band structure with spherical electron and hole pockets of unequal radii and a "nonmagnetic" reservoir is examined. A first-order commensurate-incommensurate transition is found, but the paramagnetic transition remains second order. The influence of generalized Kohn anomalies on the transition is investigated, and it is found that though they lead to anomalous behavior in the limit of vanishing antiferromagnetism, there is no change in the order of the transition.

I. INTRODUCTION

In recent years, the antiferromagnetism of Cr and its alloys has been studied extensively. It has been well established that the antiferromagnetism in Cr and its alloys is itinerant in nature and is an example of an Overhauser spin density wave arising from correlations between the conduction electrons.[1] The importance of the peculiar nature