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Resistive Behavior of Dilute *Pd* Fe Alloys about the Curie Temperature[*]

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We have analyzed the behavior of the magnetic part of the electrical resistivity ρ_m for four dilute samples of *Pd*Fe(0.1, 0.25, 0.50, and 1.0 at. % of Fe) in the vicinity of the Curie temperature. The ferromagnetic critical temperature for these samples was determined by the location of the maximum in $d\rho_m/dT$ and was found to vary with concentration as c^n, with $n = 1.65 \pm 0.05$. The "singularity" of $d\rho_m/dT$ was found to be much stronger than logarithmic with the exponents varying over a wide range of values. Also, we find that particularly in one sample, $Pd_{0.995}Fe_{0.005}$, the long-range nature of the short-range spin fluctuations plays a significant role in determining the behavior of $d\rho_m/dT$.

I. INTRODUCTION

The magnetic properties of dilute magnetically ordered alloys have been a subject of extensive study in recent years. In such a study, one looks for the basic mechanism responsible for the formation of localized moments, their interaction with the matrix, and the nature of the transition to the ordered state. The understanding of the nonequilibrium properties, such as the transport coefficients, in these systems has left much to be desired. Even though widely different systems do behave fairly similarly with regard to their equilibrium properties near the critical temperature, the transport coefficients do show striking dissimilarities. These differences in the critical behavior of the transport coefficients are expected to shed a great deal of light on the various interaction mechanisms in these systems.[1]

The palladium-iron alloys have the characteristic property of forming a well-defined ferromagnetic state even for very low concentrations of iron. In fact, in the case of *Pd* Fe, there exist distinctly different regions of concentration-dependent behavior. For example, the ordering temperature increases linearly with concentration in the concentration range 1.0 to about 4 at.% of iron, and in the concentration range 4–12 at.% of iron but with a slope which is smaller by a factor of about 3.[2] Above 1 at.% of Fe concentration, the palladium matrix shows an increasingly homogeneous magnetic polarization, and at about 3 at.% of iron, where the neutron-scattering cross section becomes effectively independent of the scattering vector, the polarization is uniform.[3] On the other hand, in the case of lower concentrations (1% and below), the ordering temperature varies more strongly than linearly with concentration. For concentrations less than 1 at.% of Fe the neutron-scattering data[3] have shown that the polarization of the palladium matrix extends out from an iron atom with a range of about 10 Å. This giant moment (as large as $10\ \mu_B$ for $\frac{1}{4}$% Fe) in these alloys is the result of contributions from a large number of palladium atoms in the matrix affected by a single iron atom.

Theoretical studies of various properties for these alloys have been carried out by many workers. In these studies, both spin-wave and cluster models have been used with reasonable success.[4–8] In particular, Coles and Turner,[7] and Doniach and Wohlfarth[6] have demonstrated that damped spin waves give an enhanced contribution to the specific heat, which is in fair agreement with the experimental results of Veal and Rayne.[9] Also, the studies of the temperature dependence of the electrical resistivity of dilute *Pd* Fe alloys by Williams and Loram[10] are explained on the basis of scattering contributions from the spin waves.

The critical behavior of these alloys to the ordered state is of particular interest, since, quite evidently, in this behavior the matrix plays a very significant role.[11–13] Our previous work[14] has emphasized the high-concentration regime, thereby limiting the information to be gained about the effects of the long-range part of the polarization which we expect to be more important at low concentrations. In an effort to better understand the dynamic aspect of the spin ordering in the lower-concentration range, we have studied the critical behavior of the electrical resistivity of four samples of dilute *Pd* Fe alloys in the vicinity of the ordering temperature. In Sec. II we describe the experimental procedure; in Sec. III we present the experimental results and some relevant discussion; and in Sec. IV we evaluate the critical exponents.

II. EXPERIMENTAL DETAILS

Measurements of the electrical resistivity were made on samples of *Pd* Fe with concentrations of 0.10, 0.25, 0.50, and 1.0 at.% of Fe in the temperature range 1.2–80 °K. In the case of the 0.10-at.% sample, the measurements were extended down to 0.40 °K. All samples were cut into small bars from large homogenized ingots. They were then cold rolled to about 1-mil thickness. The sample sizes used were about 2 to 3 mm wide and about 2 to 3 cm long. After rolling, the samples were etched to remove any surface contamination due to rolling and then were annealed at about 700 °C for several hours. One of the samples (0.25 at.%) was later further heat treated at 1100 °C in a sealed evacuated quartz tube for about five days and water quenched and the measurements were repeated. A standard four-point probe technique was employed to measure the electrical resistivity at any given temperature. A dc supply, which was regulated to better than 1 part in 10^5, was used to provide about 100 mA of current through the samples. The potential difference thus generated was measured with a Honeywell 2779 potentiometer along with a Keithley nV null detector thereby providing measurements to better than a few nV. The readings were taken at every temperature with the current reversed to eliminate the effects of any extraneous emf. The samples were in a good thermal contact with a large high-conductivity copper block into which a manganin heater was imbedded. In the temperature range 4.2–80 °K the copper block was in weak contact with a liquid-helium bath and its temperature was controlled with the help of an automatic temperature controller. A Ge-resistance thermometer was used to measure the temperature of the block, and the temperature was maintained constant for every reading to within a few mdeg. For temperatures below 4.2 °K, successive pumpings on liquid He^4 and liquid He^3 were used.

The data for the electrical resistivity $\rho(T)$ were recorded at various small temperature intervals, and then were computer differentiated, point by point, at every temperature, with a best five-point quadratic fit. To facilitate the critical analysis, $d^2\rho/dT^2$, the second derivative of $\rho(T)$, was also computer generated from the $d\rho/dT$ data in a similar manner.

III. RESULTS AND DISCUSSION

In the case of dilute *Pd* Fe alloys ($c_{\mathrm{Fe}} \leq 1$ at.%) one can separate various contributions to the electrical resistivity by writing it in the form

$$\rho(T,c)=\rho_m(T,c)+\rho_{\mathrm{host}}(T)+\rho_0(c)\ ,$$

where $\rho_m(T,c)$ represents the contributions from the magnetic or spin-disorder scattering mechanisms, $\rho_{\mathrm{host}}(T)$ is the temperature-dependent part of the resistivity of pure palladium host, and $\rho_0(c)$ is the residual resistivity of the alloy. We are mainly concerned in this paper with the critical behavior of the magnetic part of the resistivity ρ_m.

In Fig. 1, we present

$$\Delta\rho_m(T)=\rho_m(T)-\rho_m(T_L),$$

where T_L is the lowest temperature of the data and $d\rho_m/dT$ for two samples with concentrations of 0.1-at.% and 0.25-at.% Fe. The critical temperature T_c may be taken as the temperature where the temperature coefficient of $\rho_m(T)$, that is $d\rho_m/dT$, shows a singular behavior. In our samples, $d\rho_m/dT$ shows a maximum at a temperature which we take to be T_c. At temperatures much larger than T_c, where the spin-disorder scattering is no longer changing with temperature, we find that the magnetic contribution to the resistivity ρ_m is constant as shown in Fig. 1. As the temperature is lowered towards T_c, ρ_m decreases at first slowly owing to the increasing presence of short-range order and then sharply when the long-range order sets in. These features are clearly exhibited in the $d\rho_m/dT$ curves. In fact, it is the behavior of $d\rho_m/dT$ which better characterizes the nature of the transition. In both these samples the peak is fairly sharp. Below T_c the curve falls gently for the 0.25% sample, shows a small plateau, then falls again. Similar characteristics are observed in the case of the other two samples of 0.5 and 1.0 at.% of Fe as seen in Fig. 2. The width of the transition generally increases as the concentration is increased. In the samples of 0.25, 0.50, and 1.0 at.% of Fe the half-widths determined from the region of the curve for $T > T_c$ are approximately proportional to the concentration with values of 1, 2, and 4 °K, respectively, with the corresponding relative half-widths $\Delta T/T_c = 0.24$, 0.15, and 0.12, indicating some decrease with increasing concentration. In the 0.1% sample, the lowest concentration studied, the half-width is about 0.1 °K ($\Delta T/T_c = 0.13$). For $T > T_c$, the 0.50% sample shows a significantly different behavior. As the temperature increases, the $d\rho_m/dT$ curve falls off first rapidly and then slowly towards a shallow minimum at about 21 °K; then it rises to a broad maximum located at about 30 °K as the temperature is further increased. This behavior is definitely present in the raw $d\rho_{\mathrm{total}}/dT$ data and

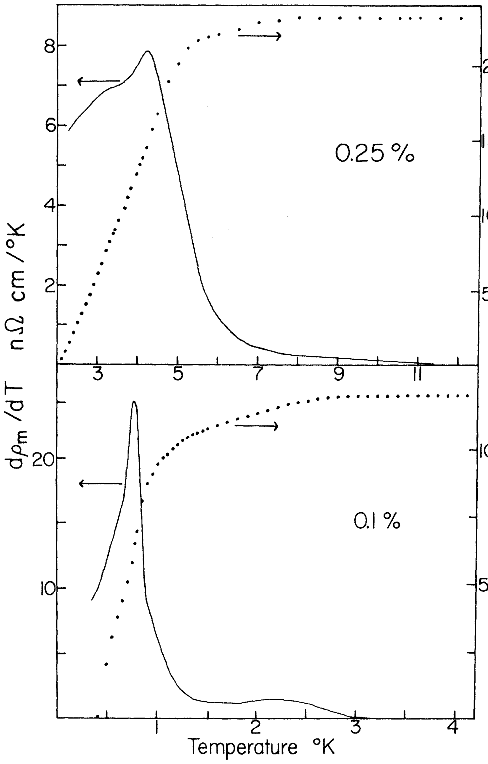

FIG. 1. $\Delta\rho_m(T)=\rho_m(T)-\rho_m(T_L)$ and $d\rho_m/dT$ for 0.1 and 0.25 at.% of Fe in Pd. T_L is the lowest temperature of the data and is 0.4 °K for 0.1% and 2.0 °K for 0.25% sample. The arrows indicate the respective ordinate scales.

not in the pure Pd data, and therefore we conclude that this behavior is to be ascribed to the presence of the iron impurity. This conclusion is reinforced by the fact that the analysis of this sample showed no significant presence of any other magnetic or nonmagnetic component. A weaker and sharper maximum can also be seen in the 1.0% sample, whereas the 0.1% sample shows a broadened region subsequent to the sharp drop in $d\rho_m/dT$, and the 0.25% sample did not show any clear evidence of such a behavior.

The general behavior of the $d\rho_m/dT$ curves is, as was recently pointed out,[15] similar to that of the specific heat.[9] Before proceeding to a quantitative description of the general features of the critical behavior common to all the samples reported in this paper, we will discuss qualitatively a few characteristics of the 0.5% sample. For $T \lesssim T_c$, the contribution to $d\rho_m/dT$ comes mainly from the onset of long-range order, whereas above T_c the behavior is mainly governed by the short-range order, its nature and the mechanism responsible for it. In this sample, the behavior of $d\rho_m/dT$ seems not unlike that predicted from the model of de Gennes and Friedel[16] and Kim.[17] In this model, the dominant contributions to $d\rho_m/dT$ arise from the long-range nature of short-range spin fluctuations, which in turn gives a sharp maximum (at T_c^-) in $d\rho_m/dT$ followed by a minimum (at T_c^+) as the temperature is increased. In our sample, the observed behavior is not exactly that predicted by the above model but there is strong evidence for some contribution from this mechanism; the total behavior of $d\rho_m/dT$ for $T > T_c$ results from a combination of the above mechanism with a contribution from direct short-range spin ordering.

It is not clear why such a behavior is most pronounced in only one of our samples ($c = 0.50\%$). Perhaps at this concentration the iron moments, along with the matrix polarization cloud, are sufficiently well separated so that we may view these as localized cluster moments. As one departs from this concentration in either direction this may no longer be true. As the concentration is increased, the iron sites come closer and the polarization clouds overlap. For decreasing concentration, the range of polarization increases,[18] thus affecting the long-range part of the short-range spin fluctuation. We may therefore expect a critical interplay between the cluster separation and the range of polarization such that the resistive behavior due to the long-range part of the short-range spin fluctuations may be strongly dependent

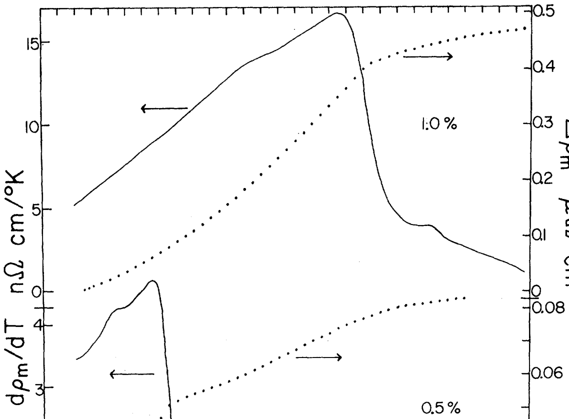

FIG. 2. $\Delta\rho_m(T)=\rho_m(T)-\rho_m(T_L)$ and $d\rho_m/dT$ for 0.50 and 1.0 at.% of Fe in Pd. T_L is the lowest temperature of the data and is 2.0 °K for 0.50% and 4.2 °K for 1.0% sample. The arrows indicate the respective ordinate scales.

upon concentration in this regime where the range of polarization is comparable to the iron-iron separation. Furthermore, the observation of some sensitivity to the variation in heat treatment of these samples supports this picture. A comprehensive study of the dependence of the effects of heat treatment on the resistive behavior in dilute systems is underway. In addition, recent measurements of the spin lattice relaxation rate of Pd^{105} show a definite peak in the relaxation rate at about this concentration ($c = 0.50\%$).[19]

The critical temperature determined from the location of the dominant maximum of $d\rho_m/dT$ varies as c^n ($n = 1.65 \pm 0.05$) in the concentration range studied, in contrast with its linear dependence at higher concentrations, which might be understood in terms of a molecular field or random-phase approximation. Recently, for an *s-d* exchange interaction between the conduction electrons and the impurity spins for dilute magnetic alloys, Thompson[20] has predicted the ordering temperature to be proportional to $c^{1/2}$. In the case of an isotropic Gaussian distribution of the matrix polarization the concentration dependence of the ordering temperature is $T_c \propto \exp(-B/\sigma^2 c^{2/3})$, where B depends upon the lattice constant and σ is the Gaussian width.[21] On the other hand, as is the case in *Pd*Fe where the polarization of the matrix plays an important role, the dependence of T_c on concentration is determined by the exact details of the matrix polarization which may not be strictly Gaussian.

A comparison of the ordering temperature obtained from different measurements for $c \lesssim 1.0$ is shown in the Fig. 3. In view of the complexities of deducing an accurate value of the transition temperature from the variety of experiments referred to in Fig. 3, we will not consider further the differences in the various transition temperatures.

IV. CRITICAL EXPONENTS

In order to characterize the critical behavior of $d\rho_m/dT$ we write it in the form[22]

$$\frac{d\rho_m}{dT} = \frac{A}{\lambda}(\epsilon^{-\lambda} - 1) + B,$$

where A and B are constants, $\epsilon \equiv |(T - T_c)/T_c|$, and λ is the critical exponent, which if it were zero would indicate a logarithmic singularity. To facilitate the analysis, we take the temperature derivative of $d\rho_m/dT$:

$$\frac{d^2\rho_m}{dT^2} = -\frac{A}{T_c}\epsilon^{-(\lambda+1)},$$

and then log-log plots would directly give us the values of λ. It may be added here that this type of log-log plot is a much more sensitive test for the nature of the singularity rather than a semilog plot of $d\rho_m/dT$ versus $T-T_c$, as is sometimes used.[23] In Fig. 4 we have shown the log-log plots of $d^2\rho_m/dT^2$ versus ϵ for $T > T_c$. As is evident from the curves, there is no evidence of a logarithmic singularity[24] (corresponding to a slope of -1 or $\lambda = 0$) in our results. In fact, as ϵ increases each curve rises towards a maximum and then falls off. Below the temperature corresponding to the maximum (or for small ϵ) the precise nature of the curve depends sensitively on the exact choice of T_c. For larger values of ϵ beyond the maximum where the behavior is not sensitively dependent upon the exact choice of T_c, we find the values of λ to be about $+5$ for 0.50 and 1.0% Fe, and about $+3.5$ for 0.25% Fe. For the sample of 0.1% Fe, the behavior for large ϵ is not simple and the data yield a $\lambda \sim +0.3$ for ϵ up to 0.35 and about $+2$ for higher values of ϵ.

The presence of a maximum in $-d^2\rho_m/dT^2$ which occurs in the neighborhood of $\epsilon \sim 0.1$ might be understood in terms of spin clusters of polarization associated with each impurity center. As the temperature ($T > T_c$) is lowered towards T_c, the size of clusters would grow thereby giving increasingly smaller contribution to the magnetic part of the

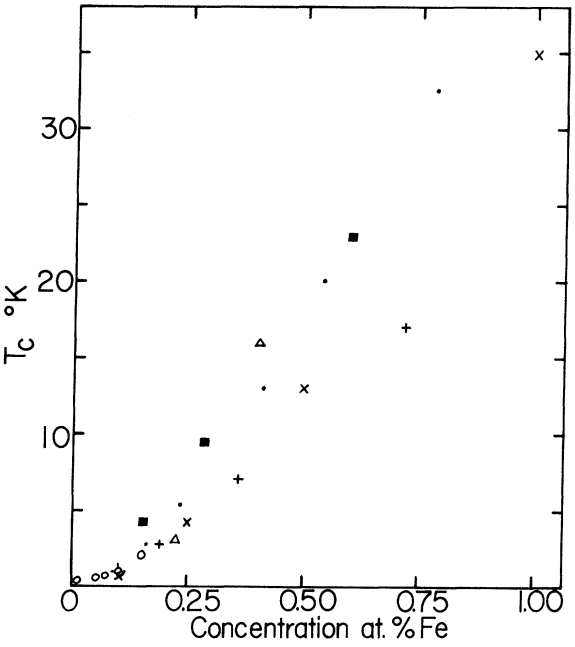

FIG. 3. The critical temperature T_c versus concentration for $c \leq 1.0$ at. % of Fe in Pd. ×: the present data; •: Williams and Loram, Ref. 10; ○: M. McDougald and A. J. Manuel, J. Appl. Phys. 39, 961 (1968); ■: J. Crangle and W. R. Scott, J. Appl. Phys. 36, 921 (1965); Δ: Trousdale *et al.*, Refs. 21 and 11; +: Veal and Rayne, Ref. 9.

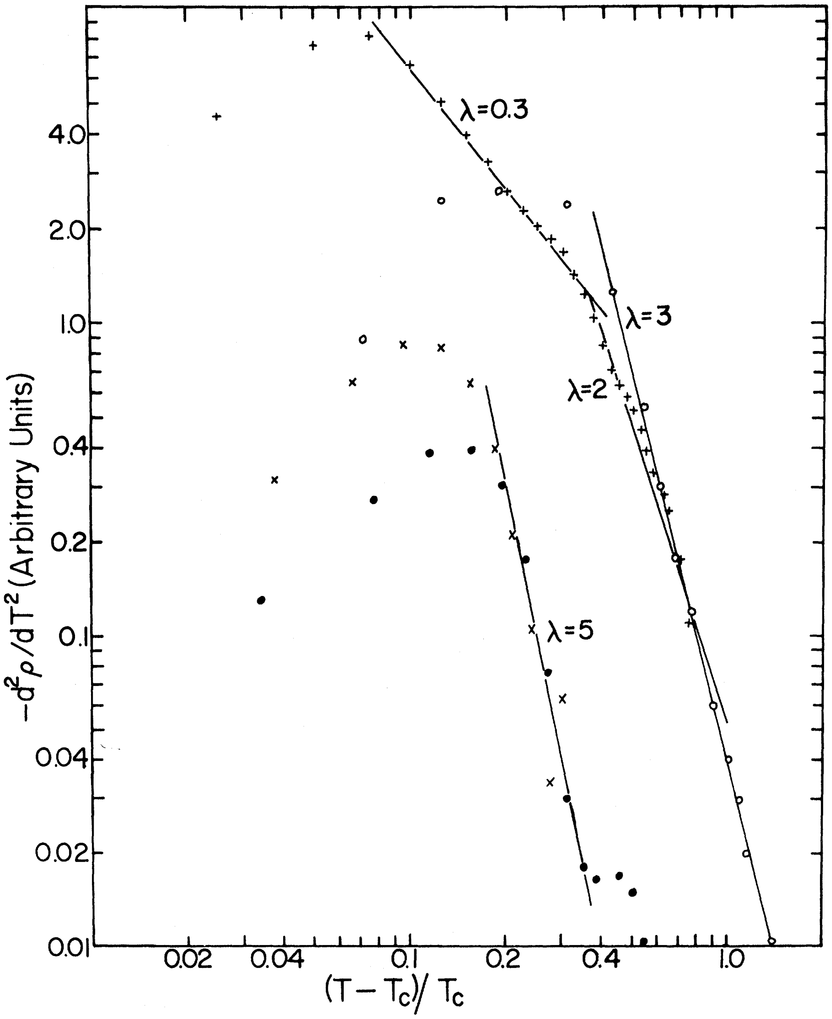

FIG. 4. $-d^2\rho_m/dT^2$ versus reduced temperature interval $\epsilon = (T - T_c)/T_c$ for $T > T_c$. +: for 0.1%; ○: for 0.25%; ●: for 0.50%; ×: for 1.0% sample. λ is the critical exponent for $d\rho_m/dT$ above T_c and is obtained directly from the slopes of solid lines.

electrical resistivity due the short-range spin ordering. As the temperature is lowered further, the magnetic clusters would coalesce and bring about a significant change in the rate of decrease of ρ_m with decreasing temperature. The maximum in $-d^2\rho_m/dT^2$ could be the result of such a behavior as well as depending somewhat on the precise choice of T_c.

On the basis of the present data and that previously available[2,14] for higher concentrations, the dependence of λ on concentration is not easily understood in terms of existing models. In addition, recent theoretical work questions the applicability of scaling laws to the critical behavior of alloys.[25] To some extent, this question arises from the difficulty of determining a well-defined ordering temperature for a random alloy. In fact, this difficulty restricts an unambiguous analysis of our data to relatively large values of ϵ and thus limits the information available about the critical region of very small ϵ. Further exploration of this problem, both theoretically and experimentally, would be valuable for the system *Pd*Fe as well as for similar highly polarizable alloy systems. This is especially interesting in a system where the range of polarization is strongly concentration dependent.[18]

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Transient Space-Charge-Limited Currents in Photoconductor-Dielectric Structures

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The theory for transient space-charge-limited currents in photoconductor-dielectric structures is presented. The analysis gives the transient current density and voltage across the photoconductor as a function of time for various values of α. Here α is a parameter that characterizes the photoconductor-dielectric structure and depends upon the relative thicknesses and dielectric constants of the two regions. In two limiting cases, our results reduce to the direct-contact (no dielectric) and open-circuit (dielectric of infinite thickness) configurations already discussed in the literature. Our rigorous mathematical expressions, involving integrals of the exponential integral, differ somewhat from the original work on the direct-contact case, although numerical results are essentially the same. The general theory presented here will broaden the spectrum of techniques already available for determining drift mobilities in insulating solids, and the relative merits of various photoconductor-dielectric geometries are discussed.

I. INTRODUCTION

In 1962 the theory for transient space-charge-limited currents (TSCLC) in insulating materials was derived independently by Many and Rakavy[1] and by Helfrich and Mark.[2] Since then the theory has found wide application in the experimental determination of drift mobilities in a large number of materials, including anthracene,[3] iodine,[4] arsenic sulfide,[5] phthalocyanine,[6] and sulfur.[7] Most of these materials form molecular solids in which the carrier mobilities are low ($\lesssim 1\ \mathrm{cm^2/V\,sec}$). For such low mobilities, Hall-effect measurements are impractical and the TSCLC technique has proved extremely valuable.

There are two major assumptions in the original TSCLC theory that impose limitations on experi-