Electrical resistivity of *Au* Fe alloys in the spin-glass, mictomagnetic, and ferromagnetic regimes

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Measurements are presented of the electrical resistivity for a series of *Au* Fe alloys with concentrations between 0.5- and 22-at.% Fe, in the temperature range 0.5 – 300 K. We have called the concentration range between about 0.5- and 8-at.% Fe, the spin-glass regime. Here we find that the impurity resistivity $\Delta\rho$ has a $T^{3/2}$ dependence down to the lowest temperatures of measurement, the coefficient of this dependence decreasing very slowly with concentration. At higher temperatures, around the "freezing" temperature T_0, the impurity resistivity is increasing linearly with temperature, and this is followed, at much larger temperatures, by a very broad resistance maximum. We have called the concentration range above $\simeq$10-at.% Fe, the mictomagnetic regime which is characterized by having large magnetic clusters and a sensitivity to thermal and magnetic history. Upon further increasing the concentration to the percolation limit $c \gtrsim 15$ at.%, such that there is sufficient overlapping among these magnetic clusters, *Au* Fe gradually develops a long-range inhomogeneous ferromagnetic regime. Again we observe a $T^{3/2}$ temperature dependence throughout both of these regimes at low temperatures, but at higher temperatures the deviation away from this dependence is much more complicated than in the spin-glass regime. Further, the onset of magnetic ordering is clearly seen in $\Delta\rho$. We have also examined the temperature dependence of the derivative of the impurity resistivity $d(\Delta\rho)/dT$, and find that throughout our whole concentration range there is a well-defined maximum which correlates fairly well with T_0. The experimental and theoretical background of these measurements is fully discussed.

I. INTRODUCTION

At the present time there is a considerable interest in "more concentrated" magnetic alloys.[1] These systems have a much larger concentration of localized magnetic impurities than the well-studied Kondo or dilute noninteracting limit,[2] and the impurities are randomly distributed at substitutional sites in the crystalline nonmagnetic host. The terms "spin-glass"[3] or "mictomagnet"[4] are now being used to describe the properties of such interacting impurity systems. Spin glasses exhibit quite strange magnetic[5] and transport[6] properties, and in this paper we would like to examine the magnetic contribution to the electrical resistivity for a typical spin-glass system, namely, *Au*Fe. This system is a particularly propitious choice since it possesses "good" Fe moments, i.e., the Kondo temperature is relatively low ($\simeq$ 0.2 K). In addition, *Au*Fe has a rather favorable solubility: of the order of 10-at.% Fe can easily be dissolved in the fcc Au matrix. Although there have been numerous resistivity measurements in this system, over many years, there has, as yet, been no systematic investigation of *Au*Fe from the spin-glass point of view.

In this paper, we will consider the lower concentration range $c \simeq 0.5$ to 8-at.% Fe in Au, as the spin-glass regime. Then, as the concentration increases ($\simeq$ 10 at.%), and there is a greater probability towards large clustering, we use the term mixed or mictomagnetic to describe the effects which dominate. Finally, for $c \gtrsim 15$ at.%, as the percolation limit[7] is exceeded, an inhomogeneous long-range ferromagnetic ordering exists. Our measurements not only include the magnetic resistivity $\Delta\rho(T)$, but also, in an attempt to characterize the "freezing" or ordering of the spins, the temperature derivative $d(\Delta\rho)/dT$. Section II contains a survey of the previous resistivity studies of the *Au*Fe system, and also the existing theories of such interacting impurity systems. In Sec. III we briefly describe our experimental technique. Section IV considers the resistivity of pure gold, which is required to determine the magnetic contribution of the alloys. In Secs. V and VI we

present our data, respectively, for the lower-concentration spin-glass regime, and for the mictomagnetic and ferromagnetic regimes. The conclusions of our work are discussed in Sec. VII.

II. PREVIOUS RESISTIVITY STUDIES OF *Au*Fe AND EXISTING THEORIES

The resistivity of the *Au*Fe system has been extensively studied. A resistance anomaly was first clearly demonstrated by Gerritsen.[8] In a detailed study of very dilute alloys (less than 25-ppm Fe), Loram *et al.*[9] showed that the resistivity could be fitted to Hamann's[10] equation with a Kondo temperature T_K of 0.24 K using a spin value S of 0.8. Susceptibility measurements[11,12] gave a similar value of T_K, but a more meaningful S of 1.5. The effect of interactions between impurities on the electrical resistivity has been studied by Ford *et al.*[13] Only below a concentration of roughly 25 ppm could the Fe impurities be considered as acting in a completely isolated manner. Above this concentration it was found that extra terms in the resistivity expression were required to account for the presence of an internal-field distribution.[14] Using an expression similar to that derived by Silverstein[15] and by Abrikosov,[16] Ford *et al.*[13] were able to show the way in which interactions were modifying the resistivity of the isolated impurities for concentrations up to 0.1-at.% Fe. The resistivity measurements of Garbarino and Reynolds[17] are in general agreement with the above observations.

Laborde and Rhadakrishna[18] have examined alloys with concentrations less than 300 ppm, down to 36 mK. They found that below the Kondo temperature, the temperature of the resistance maximum is lower than would be expected by extrapolating from data at higher concentrations.[13] This is probably due to the "weakening" of the moments below the Kondo temperature, thereby reducing the internal field, and thus the interactions between impurities, which account for the resistance maximum.

Some early resistivity measurements by Domenicali and Christenson[19] on more concentrated alloys (0.1- to 5.1-at.% Fe), made between 4.2 and 1200 K, showed a broad maximum for the impurity resistivity around 100–300 K. Another early resistivity study of very concentrated *Au*Fe alloys, up to 40-at.% Fe, was carried out by Sundahl *et al.*[20] Here there was no attempt to determine the magnetic contribution, and a smooth but "oppositely curved" $\rho(T)$ behavior was found, which varied with heat treatment. This anomalous curving becomes sharper with increasing Fe concentration and roughly corresponds to the region of magnetic ordering. In an attempt to better characterize the magnetic ordering, temperature derivatives $d\rho/dT$ were measured for concentrations of 2- to 22-at.% Fe, by Mydosh *et al.*[21] The maximum in $d\rho/dT$ approximately agreed with the magnetic-ordering temperature T_0, as determined from other measurements. Recently, within the framework of the spin-glass point of view, preliminary measurements of $\Delta\rho$ for *Au*Fe, were presented by Mydosh and Ford.[6]

Various theoretical studies have been made[15,16,22,23] to see how the presence of an internal-field distribution modifies the resistivity in the Kondo limit. In the most detailed of these, due to Harrison and Klein,[23] it is predicted that, well below the resistance maximum, the resistivity is varying linearly with temperature, with a slope which is roughly independent of the impurity concentration, and also that the temperature of the resistance maximum is increasing linearly with the impurity concentration for low c. In another approach to understanding the resistance maximum, Riess and Ron[24] have considered a broadening of the impurity-spin states because of Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions or nonmagnetic impurities. This lifetime broadening results in a change of scattering from elastic to inelastic, and only those electrons with energy greater than the width of the impurity state can be inelastically scattered. Thus, below a certain temperature, a portion of the scattering processes are no longer available and the resistivity falls. A recent and detailed calculation, due to Matho and Béal-Monod,[25] considered the resistivity arising from pairs of impurities coupled via the RKKY interaction. Their approach, which is in good agreement with experiment, leads to the occurrence of a resistance maximum, and is based within the framework of the molecular-field theory.[14] However, extensions from this internal-field-distribution approximation are as yet unable to explain the details of the resistance behavior in the spin glass ($c \gtrsim 0.5$ at.%) and higher-concentration regimes.

By contrast, the calculations[26] for the long-range-ordered inhomogeneous ferromagnets (for example, *Pd*Fe, *Pd*Mn, *Pd*Co[27]) provide a favorable theoretical comparison with our experiments at low temperatures (initial $T^{3/2}$ dependence) and in the region near the spin-glass freezing temperature (linear dependence). Nevertheless, this attempted analogy may not be completely valid for the peculiar type of ordering found in spin glasses. Furthermore, the high-temperature behavior of *Au*Fe is in no sense comparable with the spin-disorder scattering found in these strongly ferromagnetic systems. Very recently, calculations[28,29] were carried out for the high-temperature resistivity of nearly magnetic metals (spin-fluctuation systems), but for good-moment alloys there seems to be little theoretical guidance, except that of a

TABLE I. Pure-gold resistivity.

T(K)	$\rho(\mu\Omega\,\mathrm{cm})$	T(K)	$\rho(\mu\Omega\,\mathrm{cm})$	T(K)	$\rho(\mu\Omega\,\mathrm{cm})$
10	0.000 50	70	0.3725	190	1.365
12	0.001 15	75	0.4155	195	1.405
14	0.002 28	80	0.4585	200	1.446
16	0.004 22	85	0.5020	205	1.488
18	0.007 20	90	0.5450	210	1.528
20	0.010 93	95	0.5870	215	1.568
22	0.0162	100	0.6295	220	1.610
24	0.0228	105	0.6715	225	1.650
26	0.0304	110	0.7140	230	1.690
28	0.0395	115	0.7560	235	1.730
30	0.0497	120	0.7965	240	1.770
32	0.0610	125	0.8370	245	1.812
34	0.0733	130	0.8780	250	1.852
36	0.0872	135	0.9190	255	1.892
38	0.1012	140	0.9600	260	1.934
40	0.1152	145	1.0010	265	1.975
42	0.1300	150	1.041	270	2.015
44	0.1465	155	1.082	273.2	2.038
46	0.1630	160	1.122	275	2.056
48	0.1795	165	1.163	280	2.096
50	0.1960	170	1.204	285	2.136
55	0.2400	175	1.244	290	2.178
60	0.2840	180	1.285		
65	0.3290	185	1.325		

constant spin-disorder resistivity above the ordering temperature.[30]

It is hoped that the systematic experimental data presented in this paper will stimulate a further theoretical effort into this rather complicated, yet interesting, area of concentrated magnetic alloys. Perhaps the recently proposed spin-diffusion theory of Rivier and Adkins[31] will be an important step in this direction.

III. EXPERIMENTAL TECHNIQUES

The samples used in these resistivity measurements were made through arc melting the highest-available-purity gold and iron, and drawn or rolled into wires of foils with geometries most suitable for resistivity measurements. Each concentration was separately annealed at about 900° C for a period of a few hours in either hydrogen or vacuum, and quenched into water. Immediately after quenching, the samples were stored at liquid-nitrogen temperature until measured. It was expected that this heat treatment would provide a random distribution of magnetic impurities, at least for $c < 12$-at. % Fe.

The resistivity measurements were carried out with the standard four-point-probe potentiometric technique. An over-all voltage accuracy of better than 1 part in 10^4, with a precision of a few nanovolts, was utilized in conjunction with a dc current, maintained constant to about 1 part in 10^5. Measurements were made down to 0.5 K in a conventional He^3 resistivity cryostat, details of which have already been described.[9] Below 4.2 K, temperatures were measured with He^3 and He^4 vapor-pressure thermometers. Above 4.2 K, a constant-volume gas thermometer and a germanium thermomemeter were used, and a copper-constantan thermocouple was employed above the liquid-nitrogen temperature. Temperatures were stabilized to within 1 mK below 4.2 K and to within 0.5% of the temperature at higher temperatures. The uncertainty in temperature below 4.2 K did not exceed a few millidegrees; above 4.2 K, it was always known to within 0.5% of the given temperature. The sample dimensions were determined by direct measurement to an accuracy of ±2%, and by a length measurement and weighing procedure to an accuracy of within ±0.5%. As described in Sec. IV, the resistivity of pure gold was subtracted at each temperature to determine $\Delta\rho$ ($\Delta\rho = \rho_{\text{alloy}} - \rho_{\text{pure gold}}$), which we take to be the magnetic or spin resistivity. These data, which were obtained at rather close temperature intervals $\Delta T \simeq 1$ K, were point-by-point computer fitted over a five-point span to a best-fit second-order polynomial, and then point-by-point computer differentiated to give $d(\Delta\rho)/dT$.

IV. PURE-GOLD RESISTIVITY

In the course of our work, we have measured the resistivity of different samples of pure gold as a function of temperature on many occasions, and these data are summarized in Table I. Although the resistivity of pure gold has frequently been measured, the data are rather scattered in the literature, and the resistivity values are often only tabulated at rather widely spaced temperature intervals.[32] In Table I, the residual resistivity, equal to the resistivity at 4 K, has been subtracted out. At 273 K, we measured 2.04 $\mu\Omega$ cm for the resistivity of pure gold, whereas White and Woods[33] quote 2.01 $\mu\Omega$ cm, and Gerritsen[34] quotes 2.02 and 2.04 $\mu\Omega$ cm for two specimens. These differences can be accounted for by a 1% error in the sample dimensions. Damon *et al.*[35] tabulate data for the pure-gold resistivity up to 500 K and Domenicali and Christenson[19] show graphs up to 1200 K.

At low temperatures, the subtraction of the pure-gold resistivity from the alloy resistivity gives the magnetic or spin resistivity. However, at higher temperatures, where phonon scattering becomes significant, such a way of obtaining the magnetic resistivity becomes increasingly uncertain, because Matthiessen's rule is not well obeyed. Deviations from Matthiessen's rule have been extensively studied, and much of this work has been summarized in the review article by Bass,[36] who has shown that there are numerous mechanisms which can give rise to these effects. In a recent paper, Whall *et al.*[37] have examined the breakdown

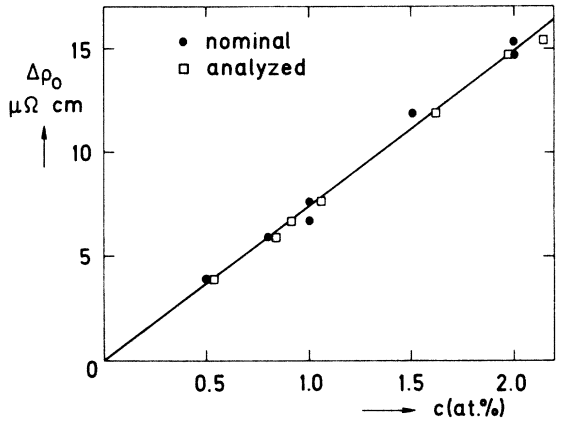

FIG. 1. Residual resistivity $\Delta\rho_0$ ($\mu\Omega$ cm) vs concentration c (at.%) for some of the more dilute *Au*Fe alloys studied. ○: nominal concentration; □: analyzed concentration.

of Matthiessen's rule in gold-based alloys containing 3*d*-transition-metal impurities, but for much lower concentration than those considered in the present paper. It was found that at low temperatures the deviation was proportional to the phonon resistivity, and the results were interpreted in terms of the differing anisotropies of the relaxation times for phonon and impurity scattering over the Fermi surface. Such an interpretation has been used extensively in discussing deviations from Matthiessen's rule.[38]

In many alloy systems including the one under consideration in this paper, it is extremely difficult to ascertain to what extent observed temperature dependences are being modified by deviations from Matthiessen's rule (DMR), and discussion of this point is, of necessity, speculative.[39] Nevertheless, past work may serve as a useful guide. Efforts to predict the maximum possible magnitude of the DMR in the present alloys, from measurements on more dilute *Au*Fe alloys[37] ($c < 0.1$ at. %), lead to the following suggestions:

(a) In the spin-glass regime, the DMR Δ is significant above about 50 K and in the case of the 1- and 2-at. % alloys can account for almost the entire change in $\Delta\rho$ over the temperature range 50–300 K. However, the extrapolated (to these higher concentrations) temperature dependence of Δ exhibits a slowly rising region with increasing temperature from 100 to 300 K. This indicates that when DMR corrections are included, there would be a stronger fall off in $\Delta\rho$ (the "true" magnetic contribution) at higher temperatures than will be illustrated in Fig. 2. Further the maximum in $\Delta\rho$ would be shifted to a somewhat lower temperature.

(b) In the mictomagnetic and ferromagnetic regimes, the maximum value of Δ is of the order of the phonon resistivity ρ_p over the entire temperature range. More likely, however, $\Delta \ll \rho_p$, and thus, the essential features of the resistivity behavior (to be shown in Fig. 7) are maintained when ρ_p is subtracted from $\Delta\rho$.

Unfortunately, these are only order-of-magnitude estimates of Δ. Also the calculations are based on observed dilute alloy ($c < 0.1$-at. %) behavior which may greatly differ from that of the present, more concentrated ($c \geq 0.5$-at. %), alloys. In view of these uncertainties, the DMR correction is neglected in the ensuing analysis. It is realized, of course, that this may be a serious omission in the case of our lowest concentrations over the temperature range 50–300 K. At all the other temperatures and concentrations considered, we feel that the observed behaviors are not substantially modified by DMR.

V. SPIN-GLASS REGIME

In Fig. 1 we show a plot of the residual resistivity $\rho(T \to 0) \equiv \Delta\rho_0$ for our lower-concentration *Au*Fe alloys. Here $\Delta\rho_0$ varies linearly with c with a slope of 7.4-$\mu\Omega$ cm/at. % Fe. Our curve of $\Delta\rho_0$ vs c connects very nicely with the dilute concentrations of Ford *et al.*[13] and with the higher concentrations of Sundahl *et al.*[20] For $c \gtrsim 5$ at. % Fe, there is a slower than linear variation with c, and $\Delta\rho_0$ reaches a maximum of $\simeq 55$ $\mu\Omega$ cm at about 12-at. % Fe. As c is further increased $\Delta\rho_0$ gradually levels off at $\simeq 20$ $\mu\Omega$ cm for $c \gtrsim 25$-at. % Fe. At these very high concentrations long-range ferromagnetic ordering and Fe precipitation greatly affect the residual resistivity. Figure 1 also shows the slight differences between the nominal-by weight concentrations and the analyzed compositions. In this paper we use the rounded-off nominal concentrations to designate the various *Au*Fe samples.

Figure 2 gives the full temperature dependence of $\Delta\rho$ up to 300 K for 1-, 2-, 5-, and 8-at. %-Fe alloys. The change of scale between the 1- and 2-at. % samples, and the 5- and 8-at. % samples indicates the increase of the magnetic resistivity with concentration. At low temperatures, the curves rise steeply, initially faster than T, and then practically linear with T in the region of the freezing temperature T_0 (see arrows in Fig. 2). As the temperature increases, $\Delta\rho$ reaches a very broad maximum at a much higher temperature than T_0, and, then gradually falls. This decrease in $\Delta\rho$ well above the resistance maximum can be seen very clearly in the high-temperature measurements of Domenicali and Christenson.[19]

A better indication of the low-temperature behavior for the alloys with the lowest concentrations is shown in Fig. 3, where $\Delta\rho$ is plotted from 0.5 to 25 K. The curving away from linearity at low temperatures is clearly seen, as well as the "bending" towards the slowly varying region above about 15 K.

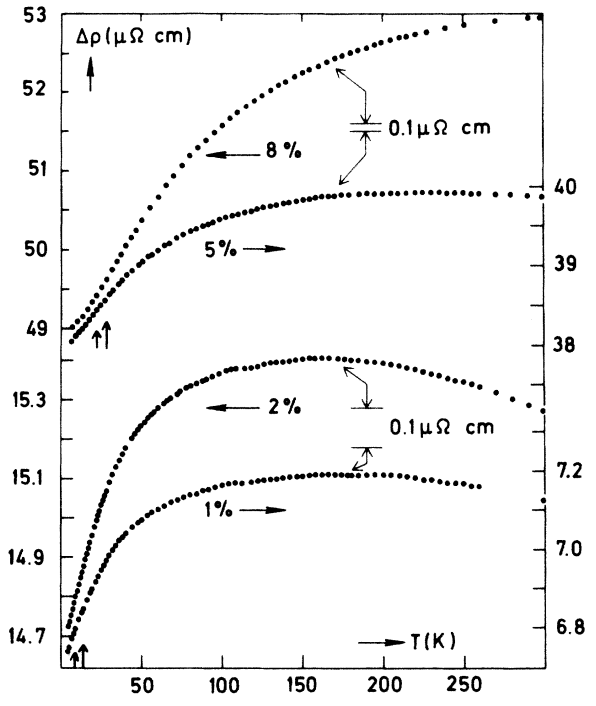

FIG. 2. Over-all temperature variation of $\Delta\rho$ ($\mu\Omega$ cm) for *Au*Fe alloys with concentrations of 1-, 2-, 5-, and 8-at.% Fe. Note the change in scale between 1- and 2-at.% alloys, and the 5- and 8-at.% alloys.

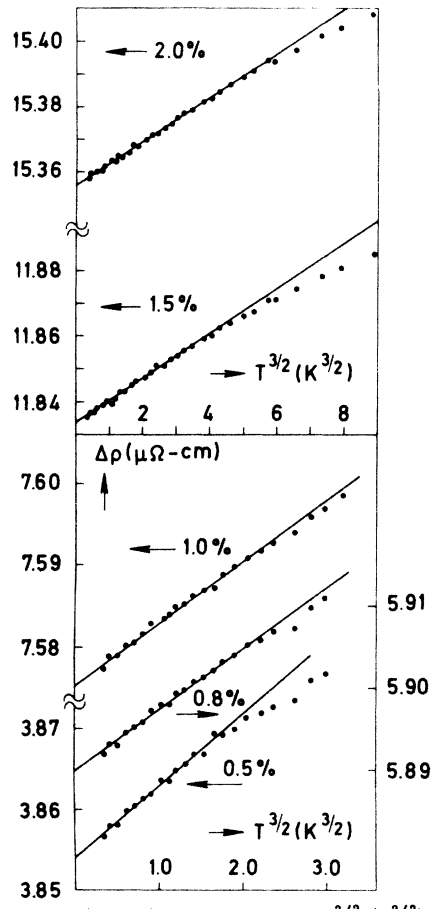

FIG. 4. $\Delta\rho$ ($\mu\Omega$ cm) plotted against $T^{3/2}$ ($K^{3/2}$) for *Au*Fe alloys in the spin-glass regime.

[The slight differences in the absolute value of $\Delta\rho$ for the 1.0- and 2.0-at.% specimens shown here and in Fig. 2 are due to the small variations of the actual analyzed compositions of two different samples (0.91- and 1.06-at.% Fe; 1.98- and 2.15-at.% Fe), which have been normalized at 1 and 2 at.%, respectively.]

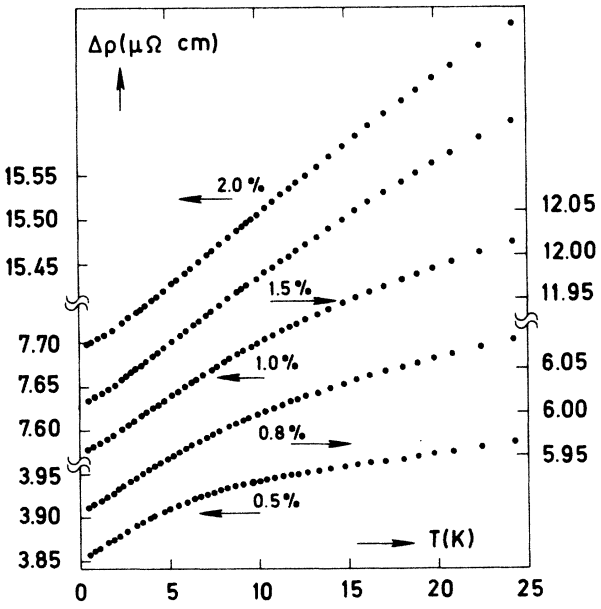

FIG. 3. Low-temperature variation of $\Delta\rho$ ($\mu\Omega$ cm) for some of the more dilute *Au*Fe alloys.

In order to examine more carefully the low-temperature initial resistivity behavior, we have plotted our data against various simple functions, and in Fig. 4 we show $\Delta\rho$ vs $T^{3/2}$. Down to the lowest temperatures of our measurements, $\simeq 0.5$ K, a best fit to the data was obtained with the simple expression $\Delta\rho = \Delta\rho_0 + AT^{3/2}$. Above 0.5 K, a T^2 fit to the data was not as effective. The range of this $T^{3/2}$ fit increases with increasing concentration, and the break away from the straight-line behavior in Fig. 4 indicates a slower than $T^{3/2}$ variation. This would represent the tendency towards a linear dependence at higher temperatures which can be seen more clearly in Fig. 3. The existence of this $T^{3/2}$ power law persists throughout the spin-glass regime. Table II summarizes the initial $\Delta\rho$ vs $T^{3/2}$ behavior for our ten spin-glass concentrations from 0.5- to 8-at.% Fe. Here the range of this simple $T^{3/2}$ dependence definitely

TABLE II. Summary of the initial $\Delta\rho$-vs-$T^{3/2}$ behavior.

c (at.% Fe)	T_0[a] (K)	Range: $T^{3/2}$ (K)	Range: $T^{3/2}/T_0$	A ($n\Omega$ cm/K$^{3/2}$)
0.5	5.5	1.2	0.22	9.6
0.8	7.5	1.2	0.16	7.8
1.0	8.5	1.6	0.19	7.7
1.5	11	2.3	0.21	7.0
2.0	14	3.0	0.21	6.8
3.0	16	6.0	0.38	6.5
4.0	20	7.0	0.35	5.5
5.0	22	12	0.55	5.5
6.5	25.5	16	0.63	5.4
8.0	28	18	0.64	5.1
12	36	40	1.1	7.5
17	130	33	0.25	17
22	220	55	0.25	22

[a] T_0 was determined from the sharp peaks in the low-field ac susceptibility measurements of Cannella and Mydosh, Ref. 5.

increases in absolute T up to 8 at. %, but there is less increase when normalized with respect to the freezing temperature: $T_{3/2}/T_0$. In addition, the coefficient of the $T^{3/2}$ dependence, A, decreases very slowly with concentration in this regime. Figure 5 gives a semilog plot of A vs c for the ten different compositions. The straight line shows that A is proportional to $-\ln c$. This implies, a very weak concentration dependence of A, and a slowly varying power law such as $c^{-1/5}$ would be an equally good fit to our data.

In an attempt to characterize, from our resistance measurements, the magnetic ordering which occurs in spin glasses, we have computer differentiated $\Delta\rho(T)$ to obtain the temperature coefficient of the magnetic resistivity: $d[\Delta\rho(T)]/dT$. Recent theories[40] and experiments[41] on a variety of magnetic materials confirm that the long-range ordering is distinguished by a maximum, or even a divergence in the case of pure magnetic elements and crystallographic ordered substances, in $d\rho/dT$. Figure 6 shows the results of such differentiations for 1, 2, 5 and 8 at. %. Each sample is characterized by a rather well-defined maximum in $d(\Delta\rho)/dT$. Even the lowest concentration 0.5- and 0.8-at. % samples exhibit maxima at very low temperatures. As can further be seen from Fig. 6, the magnetic temperature coefficient of resistivity falls off with increasing temperature, and gradually becomes negative at sufficiently high temperatures. For the 8-at. % specimen, this would probably occur above room temperature. The salient features of these and higher-concentration plots are collected in Table III. It should be noted that there seems to be a definite correlation between the "freezing" or ordering temperature T_0 and the temperature T_m at which the maximum in $d(\Delta\rho)/dT$ occurs. The agreement between these two temperatures becomes quite good for concentration above about 2 at. %. However, this is not true for the 12-at. % alloy, where, a meaningful agreement is prevented by the complex nature of the mictomagnetic state with its large clusters and strong dependence upon sample preparation and thermal history. Here the ordering temperature can be greatly varied ($\simeq$ 50 K) by various heat treatments.[42] For the 17- and 22-at. % samples, there is also good agreement between T_0 and T_m, but this is to be expected due to the long-range ferromagnetic order which becomes dominant at these concentrations.

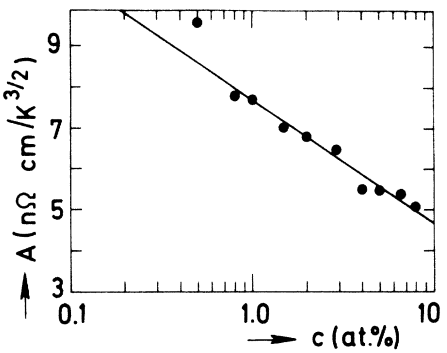

FIG. 5. Coefficient of the $T^{3/2}$ dependence A vs $\log_{10}c$.

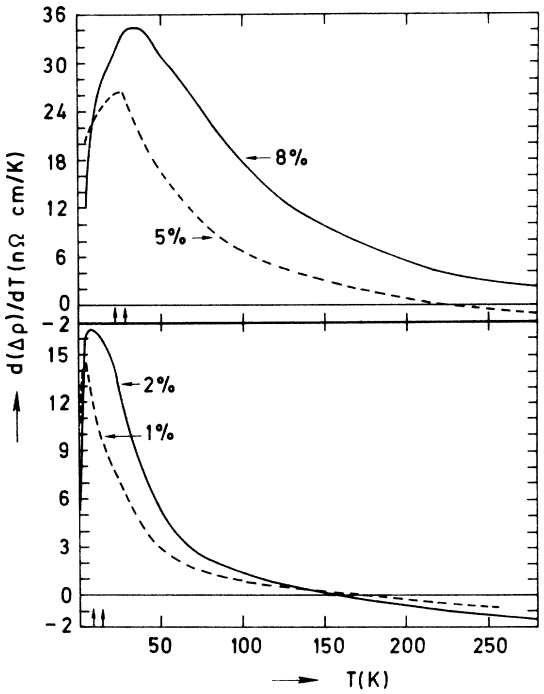

FIG. 6. Temperature dependence of the temperature derivative of the impurity resistivity $d(\Delta\rho)/dT$ ($n\Omega$ cm/K), for AuFe alloys with concentrations of 1-, 2-, 5-, and 8-at. % Fe.

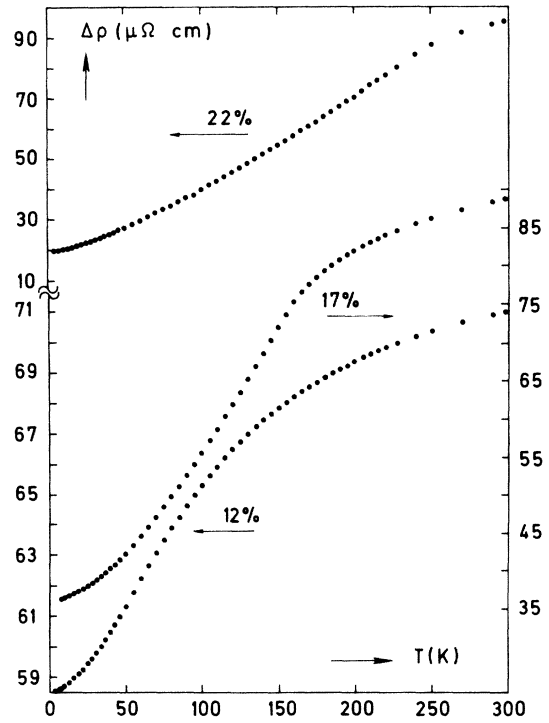

FIG. 7. Over-all temperature variation of $\Delta\rho$ ($\mu\Omega$ cm) for *Au*Fe alloys with concentrations of 12-, 17-, and 22-at.% Fe. Note the different scales used for each alloy.

TABLE III. Summary of $d(\Delta\rho)/dT$ behavior.

c (at.% Fe)	T_0 (K)	T_m (K)	T_μ (K)
0.5	5.5	1.6 ± 1	?
0.8	7.5	2.8 ± 1.5	?
1.0	8.5	4.0 ± 2	170
1.5	11	6.5 ± 3	?
2.0	14	8.0 ± 4	165
3.0	16	14 ± 5	140
4.0	20	18 ± 5	165
5.0	22	25 ± 5	225
6.5	25.5	27 ± 5	$\simeq$300
8.0	28	34 ± 10	>300
12	36	55 ± 10	>300
17	130	137 ± 10	>300
22	220	212 ± 10	>300

VI. MICTOMAGNETIC AND FERROMAGNETIC REGIMES

We now consider the higher concentration alloys, 12-, 17-, and 22-at.% Fe, which we designate as mictomagnetic for the lower concentration end, and ferromagnetic at the upper limit. There is no sharp transition between these various regimes of behavior, only a gradual growth of cluster size until there is sufficient overlap to cause ferromagnetism. Figure 7 shows the over-all temperature behavior of $\Delta\rho$ for these three concentrations. Here the magnetic contribution is rather large with the room temperature resistivity increasing with concentration while $\Delta\rho_0$ decreases. The effect of magnetic ordering is clearly indicated (especially for 17- and 22-at.% alloys) by the "knee" in the $\Delta\rho(T)$ curves. There is a spin-disorder scattering of roughly 9, 38, and 43 $\mu\Omega$ cm for 12, 17, and 22-at.%, respectively. At low temperatures the curvature towards a higher power in T (stronger than linear) is found. An attempt to fit this initial resistivity from $\simeq 5$ K upwards to a simple power law resulted once more in the expression $\Delta\rho = \Delta\rho_0 + AT^{3/2}$ (see Fig. 8). However, here the coefficient A is larger, and greatly increases into the ferromagnetic concentrations. Further, as can be seen from Fig. 8, the details of the three alloys differ among themselves. The 12-at.% sample follows a $\Delta\rho \propto T^{3/2}$ law almost up to its ordering temperature; then with increasing temperature, there is a slight deviation towards a higher power law, and finally towards a linear dependence. For the 17-at.% concentration, in a limited region well below T_0, $\Delta\rho$ follows a $T^{3/2}$ dependence, but de-

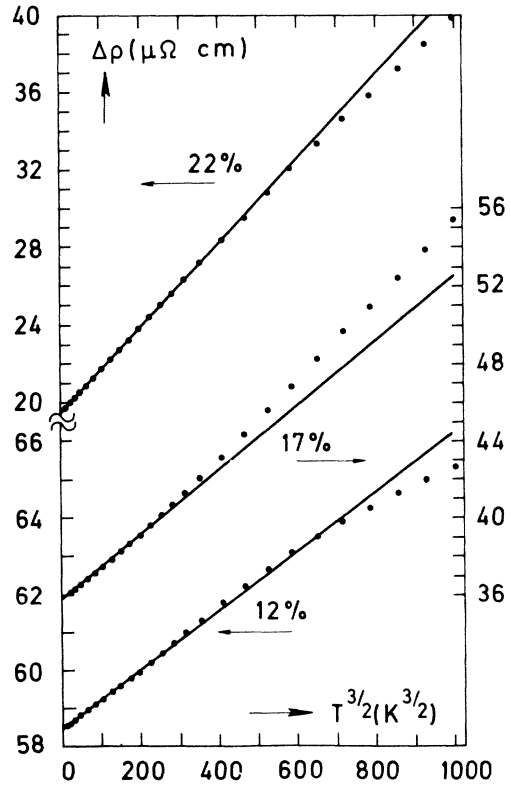

FIG. 8. $\Delta\rho$ ($\mu\Omega$cm) plotted against $T^{3/2}$ ($K^{3/2}$) for *Au*Fe alloys with 12-, 17-, and 22-at.% Fe.

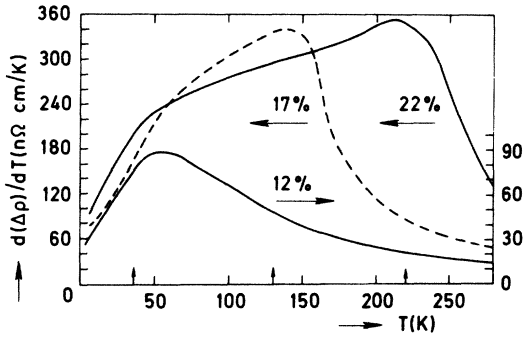

FIG. 9. Temperature dependence of the temperature derivative of the impurity resistivity $d(\Delta\rho)/dT$ ($n\Omega$ cm/K) for *Au*Fe alloys with concentrations of 12-, 17-, and 22-at.% Fe.

viates at higher temperatures towards a larger power. The 22-at.% behavior is similar to the 17 at.% at low temperatures, but then, with larger T, it changes to a roughly linear dependence. Some important parameters from the $T^{3/2}$ plots have already been given in Table II for these higher concentrations. However, we must emphasize that the lack of $\Delta\rho$ data below $\simeq 5$ K causes this initial resistivity analysis to be somewhat weak.

Now that the magnetic ordering is becoming more and more ferromagnetic in this regime, we would expect a better determination of T_0 from the $d[\Delta\rho(T)]/dT$ plots. This is presented in Fig. 9. While the changes in the temperature coefficients of the resistivity are much larger, there are no sharp peaks. However, except for the 12-at.% sample, close agreement is found between these broad maxima and T_0. Also here there is no indication up to 300 K of a negative $d(\Delta\rho)/dT$. The characteristic shapes of these curves are very much different from typical ferromagnetic systems such as *Pd*Fe[43] and *Pt*Fe.[44] Finally, it should be noted that the magnetic properties, for such an unfavorable concentration regime, above the low-temperature solubility limit, are very sensitive to the heat treatment and thermal history of the specimens, and chemical clustering of Fe atoms can readily occur.

VII. DISCUSSION AND CONCLUSIONS

It is difficult to interpret these resistivity results without the aid of the many different types of measurements performed on *Au*Fe alloys. So, wherever possible, we will try and incorporate these other conclusions as a guide to understanding our present resistivity data. Even for the lowest concentrations, we can definitely separate a temperature dependence of the magnetic resistivity $\Delta\rho(T)$ from the rather large residual component $\Delta\rho_0$. The magnitude of $\Delta\rho_0$ (7.4 $\mu\Omega$ cm/at.%) indicates a strong type of resonant scattering at $T=0$, but additionally, the random freezing of the spin glass will make another contribution to $\Delta\rho_0$. This would be a type of magnetic defect or dislocation which for $T \ll T_0$ gives a constant scattering. As the concentration is reduced, the magnetic-defect contribution to $\Delta\rho_0$ becomes less important and is overshadowed by the Kondo scattering. It is this latter contribution which keeps the value of $\Delta\rho_0$/at.% large in the dilute concentration limit. $\Delta\rho_0$ is larger for Fe impurities than for the surrounding magnetic 3*d* elements, i.e., Cr, Mn, Co, Ni. (Only with V or Ti in Au, which are large resonant scatters, is $\Delta\rho_0$/at.% greater than for Fe.)

Starting with the lower-concentration spin-glass regime, $0.5 \leq c \leq 8$ at.% Fe, the initial temperature dependence of $\Delta\rho(T)$ for $T > 0.5$ K fits very well to a simple $T^{3/2}$ power law, and then, as the temperature is increased, deviates directly towards a linear dependence (see Fig. 4). One could of course attempt fitting to a more complicated polynominal or another power law could exist at very low temperatures, but down to $\simeq 0.5$ K there is no indication of the latter in our concentration regime.[45] Therefore, we feel that this $T^{3/2}$ relationship adequately describes the data and awaits theoretical guidance or confirmation. The temperature range of the fit to this $T^{3/2}$ dependence slowly increases with concentration in absolute units, but when normalized with respect to the ordering temperature $T_{3/2}/T_0$, the range is relatively constant until 2 at.%, after which it increases (see Table II). The coefficient of $T^{3/2}$, A, very slowly decreases with concentration, varying as $-\ln c$ or $c^{-1/5}$. These initial resistivity characteristics are somewhat peculiar, and we phenomenologically attribute them to the scattering of conduction electrons by localized or highly damped spin waves within the frozen clusters, or on the periphery between clusters. At present the closest theoretical approach to our measurements is the spin-diffusion model of Rivier and Adkins.[31] Since a spin glass is characterized by a lack of well-defined long-range order, any spin-flip excitation on an impurity site is unable to propagate like a magnon, but dies away with some diffusion constant Λ. From such a starting point, Rivier and Adkins have shown that the initial temperature dependence of the resistivity increases as $(T/\Lambda)^{3/2}$. However, in its present form, the theory does have difficulty in accounting for all of the salient experimental results.

There seems to be a similarity between our observed $\Delta\rho$ behavior for *Au*Fe in the spin-glass regime, and that observed for the deviations from Matthiessen's rule, Δ, in aluminium-based alloys.[46] Here it was observed that Δ was proportional to $T^3 \ln\rho_0$, where ρ_0, the impurity resistivity, is roughly proportional to the impurity concen-

tration. The range of this T^3 temperature dependence decreased with impurity concentration. This behavior was interpreted[47] in terms of the relaxation of the conservation of momentum requirement in the electron-phonon scattering brought about by the loss of translational symmetry in an impure metal, in a similar manner to that applied for the electron-magnon scattering, which accounted for the $T^{3/2}$ temperature dependence of the dilute ferromagnet.[26] It may be that the low-temperature behavior of the deviations from Matthiessen's rule in aluminium-based alloys and also the low-temperature resistive behavior of dilute ferromagnets and spin glasses all represent different aspects of a general class of problems involving the loss of translational symmetry in an impure metal.

If we now consider higher temperatures, we observe that $\Delta\rho$ rapidly rises with an approximately linear temperature dependence. The slope of this dependence increases with concentration. This temperature region which spans T_0 is the most dynamic for spin glasses and we would expect the greatest changes in $\Delta\rho$. Here there would be a large scattering probability associated with the strongly interacting clusters, and, as the clusters randomly freeze out, a significant decrease in $\Delta\rho$. Since this is the region of "freezing," a better way to study the resistive behavior is with its temperature coefficient $d(\Delta\rho)/dT$. We have given such plots in Figs. 6 and 9, and the results are summarized in Table III. It should be noted that for the lower end of the spin-glass concentrations, the maximum T_m in $d(\Delta\rho)/dT$ occurs at a temperature which is much lower than T_0. This would indicate that a rather strange type of "ordering" is taking place which is not the usual long-range type of magnetism. Also the effect of "freezing" dramatically illustrated in susceptibility[5] and Mössbauer measurements[48] is not strongly reflected in either $\Delta\rho(T)$ or $d[\Delta\rho(T)]/dT$, e.g., the broad temperature dependences and small comparable magnitudes of these latter quantities, but that the excitations at very low temperatures, and not the freezing process, play the dominant role in determining the resistivity characteristics.

As the concentration is increased $c \gtrsim 3$ at. % Fe, there is good agreement between T_0 and T_m. Here the ordering becomes stronger with c—the curving in $\Delta\rho$ is clear and the maximum in $d(\Delta\rho)/dT$ has grown in magnitude. That the "freezing" is now directly correlated with the temperature derivative of resistivity is a possible illustration of a cooperative phenomenon in these alloys. However, the sharpness of the $d(\Delta\rho)/dT$ maxima is not particularly great, and their over-all shapes are rather strange. These properties point towards a random type of spin "freezing," and we feel that an exact description of this phenomenon is the outstanding problem in the understanding of spin-glass systems.

If we now consider yet higher temperatures, well above T_0, we observe that $\Delta\rho$ is increasing much more slowly than before. For example, regions of fit to the data could be obtained with a $\ln T$ dependence. At a very high temperature T_μ, $\Delta\rho$ gently passes through a maximum (see Table III for T_μ). For this temperature region it is rather difficult to describe the spin-glass resistivity. There was until very recently a general lack of theoretical information about resistance for cluster systems at high temperatures, e.g., the problem of CuNi.[49] Spin-fluctuation-scattering calculations for nearly magnetic systems have been carried out over wide temperature intervals by Rivier and Zlatic,[28] and Jullien *et al.*[29] The results from such theories show, in the former case, a slow $\Delta\rho$ variation ($\propto \ln T$) which becomes constant at high T, and, in the latter, a variety of resistivity maxima depending on the properties of the particular system. However, the application of such fluctuation treatments to our "good moment" *Au*Fe system is indeed questionable. In addition, the measured resistivity is difficult to analyze since the variations of $\Delta\rho$ in this temperature region are very small and are probably drastically affected by deviations from Matthiessen's rule. Further, the behavior of $\Delta\rho$ above room temperature is not known in detail. From the widely spaced high-temperature measurements of Domenicali and Christenson,[19] there is always a decrease in $\Delta\rho$ for 1.3-, 2.0-, and 5.1-at. % Fe alloys up to 1000 K. For their three concentrations, $\Delta\rho$ (1000 K) $< \Delta\rho_0$. This is a rather unusual high-temperature behavior, since apparently there is no temperature region of constant spin-disorder scattering: proportional to $c\,S(S+1)$, due to single isolated Fe impurities. Our speculation here would be that even at these very high temperatures the Fe impurities are still interacting—directly when nearest neighbors and indirectly via the RKKY at larger distances. We recall that pure-Fe metal orders at 1043 K, and that Fe in the Au matrix has a good moment ($P_{eff} = 3.25$, $T_K \simeq 0.2$ K) and can even induce some magnetic character on the Au sites.[50] So that as the Fe moments start to interact and form pairs, triplets, ..., clusters, a variety of different scattering processes, perhaps even resonant ones, become available. As a result, $\Delta\rho$ rises with decreasing T, as localized scattering from these weakly interacting spins becomes important. Perhaps some modified form of Kondo resonant scattering for multiple impurities could account for this behavior of $\Delta\rho$. With a further reduction of the temperature, these processes, due to the growth of cluster size and/or density, and the interactions between the clusters, become

slowly frozen out. Finally, as the freezing temperature is approached $\Delta\rho$ shows its largest decrease because of the cooperative or critical nature of the cluster freezing or relaxation. An alternative description of the above would be the following. The onset of local or short-range order in the Fe spins, at very high temperatures, causes a rise in $\Delta\rho$ (scattering from short-range-correlated impurities) with decreasing temperature. As the local order grows and becomes more stable, due to interactions with other regions, the spin scattering tends to be more uniform with respect to the mean free path of the conduction electrons and $\Delta\rho$ decreases. This is very sharp around T_0 [$d(\Delta\rho)/dT$ has a maximum] as the relaxation rate of cluster orientations becomes very long. Then, at the lowest temperatures, with all the local moments frozen in place, the localized excitations mainly contribute to the temperature dependence of $\Delta\rho$ and a large resonance and a magnetic "defect" scattering remain at $T = 0$ K. We await the results of theoretical calculations and of neutron-diffraction measurements to confirm these conjectures.

As the composition of Fe in the Au is increased above about 10 at. %, and the low-temperature solubility limit is exceeded, there is a tendency for very large clusters to form. This regime of giant clusters we have called "mictomagnetic" using our interpretation of the term originated by Beck.[4] It is indeed a complicated regime which depends strongly on sample preparation and heat treatments, as well as on thermal and magnetic history. With additional Fe concentration, the clusters overlap and an inhomogeneous ferromagnetic is formed. Borg *et al.*[42] showed that an enormous range of T_c's ($\simeq 50$ K) can occur with different heat treatments for a given concentration. de Mayo,[5] from a magnetization analysis as a function of heat treatment, has obtained a wide distribution of cluster sizes and densities. Thus, it is very difficult to adequately describe the effects taking place in this regime or to compare the results of the various investigations. We will try and characterize our resistivity data under the assumption of a uniform distribution of impurities resulting from our high-temperature annealing and rapid quenching. The large changes in $\Delta\rho$ for 12, 17 and 22 at. % represent a strong magnetic scattering. There is a drop in $\Delta\rho_0/c$ with c which we attribute to the ordering becoming more and more long-range ferromagnetic. Here the lattice is now without these localized "spin effects" as is the case with the random frozen alignments of the spin glass. With increasing temperature, a very large term $\Delta\rho \propto T^{3/2}$ is found, but the behavior of this dependence in the mictomagnetic regime is not the simple $T^{3/2} - T$ as can be seen from Fig. 8. The 12- and 17-at. % samples have a peculiar intermediate temperature dependence. Furthermore, the range of this behavior is anomalously large for the 12-at. % alloy and constant in $T_{3/2}/T_0$ for the 17- and 22-at. % alloys. It is very difficult here to ascertain the significance of the $T^{3/2}$ dependence. For the 17- and 22-at. % concentrations, we could perhaps invoke the theoretical treatments of Turner and Long,[26] and Mills *et al.*[26] for an inhomogeneous ferromagnet lacking translational invariance. The $T^{3/2}$ coefficient A is proportional to c for 17- and 22-at. % Fe alloys, as predicted by Mills *et al.* However, the specific differences in the $\Delta\rho$ behavior between 17 and 22 at. %, and the lack of additional concentrations prohibit any further comparison. Going on to higher temperatures, the rather steep curvature in $\Delta\rho$, particularly for 17 and 22 at. %, is similar to that found in many types of ferromagnetic systems. The "knee" just above T_0, and the maximum in $d(\Delta\rho)/T$ coinciding with T_0, all point towards a type of ferromagnetic ordering. As indicated from the more "smeared" $\Delta\rho$ character, the 12-at. % alloy would have a wide distribution of giant clusters which freeze out randomly, in zero field, without a net moment or remanence. For these three concentrations with $T > T_0$, $\Delta\rho$ continues to vary with temperature. This means the persistence of a cluster or short-range-order contribution to $\Delta\rho$. The behavior of $\Delta\rho$ at much higher temperatures is still an open question—would there be a maximum in $\Delta\rho$, followed by a decrease such that $\Delta\rho$ (1000 K) $< \Delta\rho_0$ as with the spin-glass concentrations? To generalize, for this regime, we would employ a model of scattering with giant clusters which are strongly interacting among themselves. At the lower end $\simeq 10$ at. %, $\Delta\rho$ is characterized by the clusters freezing out randomly; for the upper limit $\simeq 20$ at. %, $\Delta\rho$ manifests the behavior of an inhomogeneous ferromagnetic. Although these over-all features of the magnetic resistivity can be phenomenologically grasped in terms of such a picture, to seek a detailed understanding of the $\Delta\rho$ behavior in this regime would not be warranted, due to the complex and poorly defined metallurgical and magnetic states of the alloy system.

The rather complicated and varied behavior of $\Delta\rho$ for the 12-, 17-, and 22-at. % alloys may be related to a high-temperature inhomogeneous ferromagnetic type of ordering followed by a spin-glass freezing at a lower temperature. This would mean that the spin-glass regime persists, in a limited temperature-concentration region below a ferromagnetic regime. Thus at higher concentrations there should exist two characteristic temperatures (a Curie T_c plus T_0) which produce the anomalous deviations for the low temperature $T^{3/2}$ dependence in $\Delta\rho$ shown in Fig. 8 and also the rather broad

behavior in $d(\Delta\rho)/dT$ shown in Fig. 9.

In the present paper and in two previous papers,[9,13] resistivity measurements on *Au*Fe have been presented for concentrations ranging from under 1 ppm to 22 at.%. This range of over 10^5 in concentration probably represents one of the most detailed studies of the resistivity of any alloy system. We believe that one can distinguish five concentration regimes which gradually merge into each other. First, at very low concentrations (less than $\simeq 25$ ppm for *Au*Fe), the resistivity is dominated by the behavior of isolated impurities, i.e., the Kondo effect. As the concentration increases, interactions between impurities become important and one observes a resistance maximum. Up to a concentration of roughly 5000 ppm, there is a scaling of $\Delta\rho/c$ with T/c, also with H/c, and theories based upon a distribution of internal fields[14] are very satisfactory in describing the behavior of these alloys in this concentration range. We call this the "molecular field" or "scaling"-spin-glass regime. On further increasing the impurity concentration, the resistivity behavior is dominated by a strongly interacting set of impurities, and we designate this region simply as the spin-glass regime. Our measurements show that there is no longer scaling of $\Delta\rho/c$ with T/c, although we do find some evidence of scaling with T/T_0. The spin-glass regime persists from above about 0.5 at.% to around 10-at.% Fe. Above this concentration, where one has giant clusters and moments, we call the mictomagnetic regime. It is the most difficult region to study since the observed behavior is so complex and dependent upon the thermal and magnetic history. Finally, as the percolation limit is exceeded at around 17-at.% Fe, one has inhomogeneous long-range ferromagnetism. We wish to emphasize that none of these regions are sharply defined and each gently merges into the other. Lastly, we would like to point out that at the present time there is little general agreement on the use of the words "scaling regime," "spin glass," and "mictomagnet," and different research groups are using these terms in somewhat different contexts to those used in the present paper.[51]

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