Electrical resistivity of noble-metal-host–3d solute spin-glass alloys

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(Received 30 September 1975)

Measurements are presented of the electrical resistivity for a series of *Au*Cr, *Au*Mn, *Ag*Mn, and *Cu*Mn alloys in the temperature range 1.5–300 K and with magnetic impurity concentrations $\simeq$0.5–10 at.%. Such alloys represent typical examples of spin-glass or mictomagnetic systems. We have found that for all four systems the "impurity" resistivity $\Delta\rho$ has an initial $T^{3/2}$ dependence, the coefficient of which decreases very slowly with increasing concentration. At higher temperatures around the freezing temperature T_0, $\Delta\rho$ increases roughly linearly with T, and this is followed at much higher temperatures by a resistance maximum. This paper is a sequel to an earlier resistivity study of the *Au*Fe system. The similarities and differences between the four systems and also with *Au*Fe are described. We interpret the initial $T^{3/2}$ temperature dependence in terms of a recent spin-diffusion theory due to Rivier and Adkins. The nature of the freezing process around T_0 and its effects on $\Delta\rho$ are discussed, as well as the formation of local magnetic clusters (short-range order) at $T \gg T_0$ which give rise to the resistance maximum. The temperature dependence of the derivative of the impurity resistivity $d[\Delta\rho(T)]/dT$ has also been examined for the four systems, and it is found that $d(\Delta\rho)/dT$ has a well-defined maximum but, unlike *Au*Fe, this does not correlate well with T_0.

I. INTRODUCTION

In a previous study,[1,2] hereafter referred to as I, measurements were presented of the electrical resistivity for a series of *Au*Fe alloys over the temperature range 0.5–300 K in what we called the spin-glass, mictomagnetic, and ferromagnetic concentration regimes. The behavior of these "more concentrated" magnetic alloys is still imperfectly understood, but is currently a subject of considerable interest.[2-4] Recently, the concept, contained in the terms *spin glass*[5] and *mictomagnet*,[6] of a "freezing" of impurity spins in random directions at a well-defined temperature has been employed to describe the magnetic properties of such alloy systems. The term *spin glass* arises from certain similarities of the local-moment behavior to the molecular motions in real glasses.[7] In I the spin-glass regime was considered to be between about 0.5- and 8-at.% Fe. Here, the resistivity followed a $T^{3/2}$ temperature dependence down to the lowest temperature of measurement, the coefficient of this dependence decreasing very slowly with concentration. At higher temperatures, the impurity resistivity $\Delta\rho$ increased linearly around the "freezing temperature" T_0, and this was followed by a very broad resistance maximum. While for lower concentrations, $c \lesssim 0.5$ at.%, spin-glass effects are definitely present—the "scaling regime"—the resistivity properties are complicated by the Kondo effect and systematic measurements at very low temperatures are still required.

The initial $T^{3/2}$ temperature dependence was interpreted in terms of a recent spin-diffusion theory due to Rivier and Adkins.[8] The behavior at higher temperatures was more difficult to analyze because of the uncertain contributions from effects such as deviations from Matthiessen's rule. However, it was believed that the resistance maximum roughly corresponds to the temperature where local spin correlations exceed the thermal disorder and impurity "clusters" begin to form. Interactions between the clusters gave rise to the decrease in the resistivity at temperatures below the maximum. This was largest around T_0, because of the cooperative nature of the cluster freezing.

Above a concentration of around 10-at.% Fe, very large ferromagnetic clusters were formed at temperatures of a few hundred kelvin.[9,10] We used the term *mictomagnet*, which stems from the Greek prefix meaning "mixed," to describe the random freezing of these "giant moments" within the spin-glass matrix. As the concentration of magnetic atoms was further increased, $c \gtrsim 15$-at.% Fe, the "percolation limit" was reached, and *Au*Fe gradually showed a long-range, but inhomogeneous, ferromagnetic regime. The onset of this ordering could be clearly seen by the "knee" in the $\Delta\rho(T)$ curves for the two most concentrated alloys which were studied. In both the mictomagnetic and ferromagnetic regimes, there was also a $T^{3/2}$ initial temperature dependence,

but with the coefficient increasing with the concentration. Further the higher-temperature behavior was more complicated than that observed in the spin-glass regime. In addition to data on the temperature dependence of $\Delta\rho$, I contained data on the temperature dependence of the resistive temperature coefficient $d[\Delta\rho(T)]/dT$. With the exception of the lowest concentrations $d(\Delta\rho)/dT$ showed a well-defined maximum at a temperature which correlated quite well with T_0.

More striking than the resistivity measurements on *Au*Fe are measurements of the low-field ac susceptibility.[9,11] These show a very sharp peak at the freezing temperature T_0, enabling it to be precisely determined. The peak becomes broadened in an external field of a few hundred gauss. T_0 was found to be proportional to $c^{2/3}$, although for concentrations less than about 1-at.% Fe, T_0 tended to become more linear with c. The observation of these sharp peaks is important because it appears to be inconsistent with the early models of interacting impurities based upon distributions of internal fields. The theoretical explanation of such effects has resulted in a number of very recently proposed models and calculations.[12–16]

In view of these interesting spin-glass and mictomagnetic behaviors,[17] it was felt worthwhile to extend our resistivity investigations to other alloy systems. In order to study most simply the properties of interacting $3d$ impurities in noble-metal hosts, the alloys should satisfy two important requirements. First, it is necessary to have a high solubility of the solute impurities in the host, at least up to 10 at.%, so as to approximate the ideal case of a truly random solid solution. Second, the alloys should possess a low Kondo or spin-fluctuation temperature, $\lesssim 1$ K, so that one is dealing with "good moment" systems. An examination of the $3d$ transition-metal impurities in copper, silver, and gold show that only five systems satisfy both requirements. These are *Au*Fe and in addition *Au*Cr, *Au*Mn, *Cu*Mn, and *Ag*Mn. Many systems suffer from a very low solubility, e.g., *Cu*Fe, whereas others like *Au*V have too high a Kondo or spin-fluctuation temperature. Preliminary accounts of both the resistivity[18–20] and susceptibility[11] measurements on these five favorable systems have already appeared. The present paper is a more detailed and systematic description of the resistivity study. We will use the results of our present investigation to focus on and to discuss (a) the low-temperature excitations of the frozen spins, (b) the freezing process around T_0, and (c) the formation of local magnetic clustering at the higher temperature $T \gg T_0$.

II. EXPERIMENTAL TECHNIQUES

The resistivity of the alloys was measured between 1.5 and 300 K. The sample preparation and experimental procedures were similar to those described in I. All samples were annealed at $\simeq 900$°C for a few hours and quenched to room temperature just prior to the low-temperature measurements. The standard four-point probe technique was used; a relative accuracy of a few parts in 10^5 was obtained using a combination photoamplifier, digital voltmeter, voltage compensation technique. The resistivity of the pure host metal was subtracted from the data in all cases. Pure-gold resistivity was taken from I, pure copper from measurements of Souletie,[21] and pure silver from measurements of Kos.[22] It should be noted that this simple subtraction procedure, $\Delta\rho(T) \equiv \rho_{\text{alloy}}(T) - \rho_{\text{pure host}}(T)$, does not take into account nonmagnetic deviations from Matthiessen's rule, so that our magnetic impurity resistivity $\Delta\rho$ will be modified by such deviations especially at the higher temperatures. We shall return to this point in the following sections. The resistivity data were also point-by-point computer fitted over a five-point span to a best-fit second-order polynominal, and then point-by-point computer differentiated to give $d[\Delta\rho(T)]/dT$.

III. RESULTS

Figures 1–4 show the full temperature dependences up to $\simeq 300$ K of the impurity resistivity $\Delta\rho$ for *Au*Cr, *Au*Mn, *Ag*Mn, and *Cu*Mn. Tables I–IV summarize the important parameters for these alloys. Several features can be seen which are common to all four systems and also to *Au*Fe; see I. For instance the residual resistivity $\Delta\rho_0$ for all five systems has an initial linear dependence upon the concentration c. The slopes $\Delta\rho_0/c$ are 7.4 $\mu\Omega$cm/at.% for *Au*Fe, 4.2 $\mu\Omega$cm/at.% for *Au*Cr, 2.3 $\mu\Omega$cm/at.% for *Au*Mn, 1.5 $\mu\Omega$cm/at.% for *Ag*Mn, and 2.7 $\mu\Omega$cm/at.% for *Cu*Mn. In addition the changes of scale show that the step height $\Delta\rho(T_\mu) - \Delta\rho_0$ increases approximately linearly with concentration, indicating the increasing magnetic contribution. (T_μ is the temperature at which $\Delta\rho$ attains its maximum value.) This height is somewhat larger in *Au*Cr (0.33 $\mu\Omega$cm/at.%) and *Cu*Mn (0.28 $\mu\Omega$cm/at.%) than in *Au*Mn (0.20 $\mu\Omega$cm/at.%) and *Ag*Mn (0.18 $\mu\Omega$cm/at.%). These values are smaller than that observed for *Au*Fe (0.40 $\mu\Omega$cm/at.%). For all systems, on increasing the temperature the low-temperature resistivity rises initially faster than T, and then around the freezing temperature there is a roughly linear region in $\Delta\rho(T)$. At higher temperatures, the impurity resistivity increases slower than T and shows a

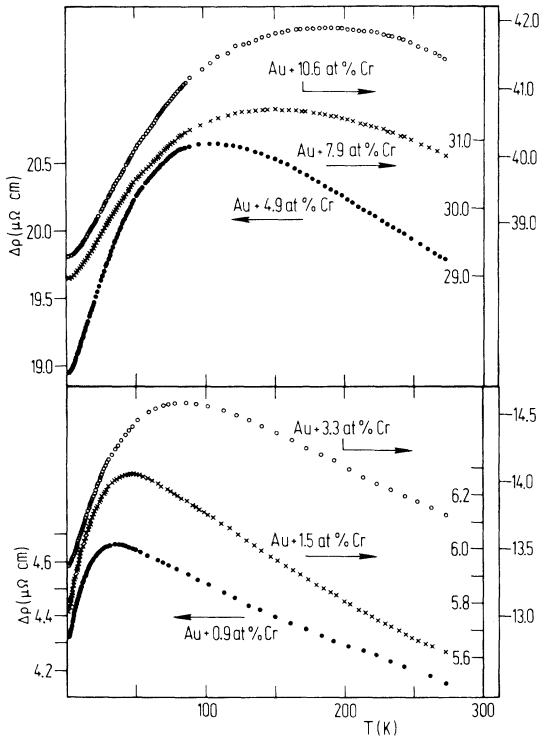

FIG. 1. Over-all temperature variation of $\Delta\rho$ ($\mu\Omega$ cm) for AuCr alloys with concentrations between 0.9- and 10.6-at.% Cr. Note the changes in scale as the concentration increases.

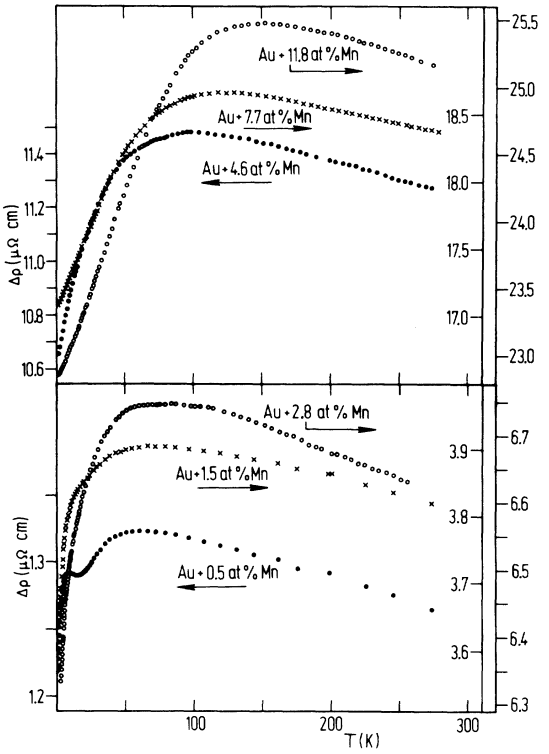

FIG. 2. Over-all temperature variation of $\Delta\rho$ ($\mu\Omega$ cm) for AuMn alloys with concentrations between 0.5- and 11.8-at.% Mn. Note the changes in scale as the concentration increases.

rather well-defined resistance maximum at a temperature T_μ before decreasing again. $\Delta\rho$ is always decreasing as room temperature is reached. Above the maximum in $\Delta\rho$ for many of the systems and concentrations studied, this decrease is surprisingly linear in T. To what extent the high-temperature data are affected by nonmagnetic contributions, such as deviations from Matthiessen's rule or changes in the normal host phonon spectrum with different impurity concentrations, is not certain, and this point will be more fully discussed in Sec. IV.

The temperature of the resistance maximum (see Tables I–IV) increases less rapidly than c, roughly between $c^{0.4}$ and $c^{0.6}$. There seems to be a parallel here between the susceptibility and the resistivity measurements. For the susceptibility[11] the peak was proportional to $c^{2/3}$ in the spin-glass regime, whereas at lower concentrations, less than $\simeq \frac{1}{2}$ at.% of impurity, in the "scaling" spin-glass regime the susceptibility maximum was proportional to c. In a similar manner, in the scaling regime, the temperature of the resistance maximum is directly proportional to c,[23–26] while the present measurements show that at higher concentrations there is a much weaker dependence.

Even though a number of general features are common to the resistivity behavior of all five systems, one can also note some differences. For example, the resisitivity maximum seems to be particularly sharp with a strong high-temperature falloff for the AuCr alloys, whereas it is extremely broad and occurs at a very high temperature for AuFe. The over-all shapes of the AuMn and AgMn alloys are very similar and represent a situation closer to AuFe, while CuMn would be more similar to AuCr. The lowest concentrations of AuMn (0.5 at.%) and AgMn (0.6 at.%) are unusual in that they show two maxima. Examination of the 0.7-at.% CuMn alloy suggests that a similar behavior would be probable at a slightly lower concentration. The data are interesting for these concentrations in that they show the occurrence of several competing processes. We believe that the first negative slope in $\Delta\rho$ observed below about 10 K is a Kondo contribution, but where the behavior of the isolated impurities is being strongly modified by interactions between impurities. As these interactions become stronger, on further decreasing the temperature, the isolated impurity spin-flip scattering process becomes increasingly suppressed to give rise to the low-temperature re-

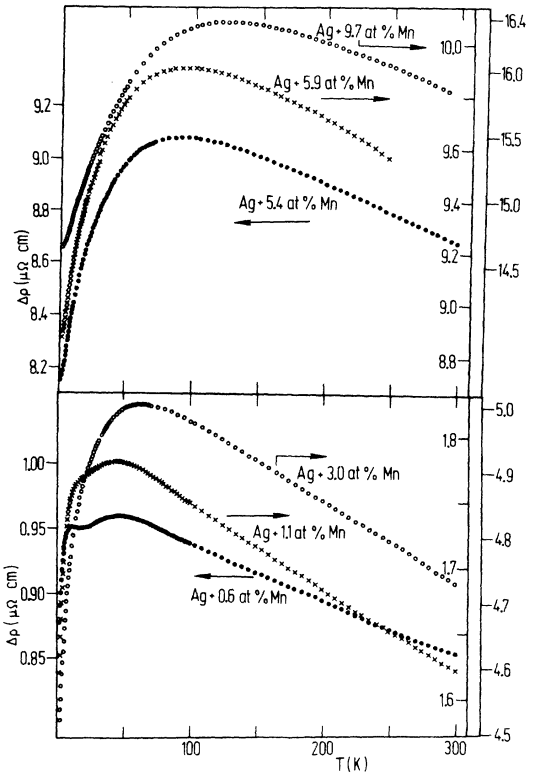

FIG. 3. Over-all temperature variation of $\Delta\rho$ ($\mu\Omega$ cm) for *Ag*Mn alloys with concentrations between 0.6- and 9.7-at.% Mn. Note the changes in scale as the concentration increases.

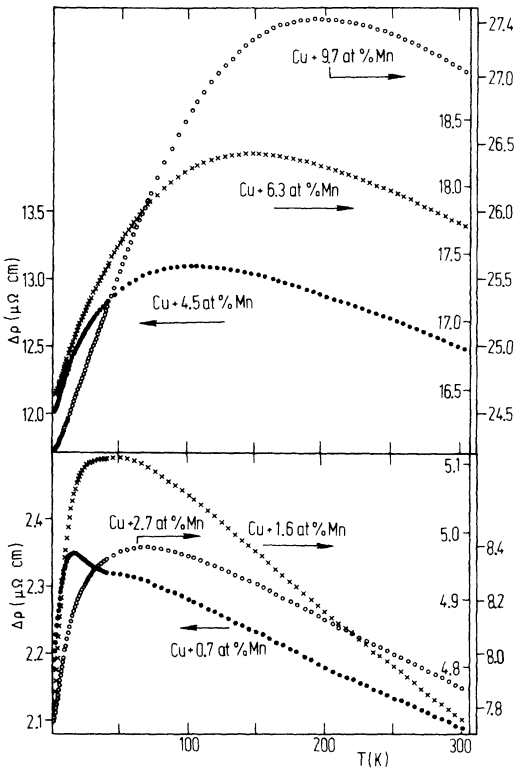

FIG. 4. Over-all temperature variation of $\Delta\rho$ ($\mu\Omega$ cm) for *Cu*Mn alloys with concentrations between 0.7- and 9.7-at.% Mn. Note the changes in scale as the concentration increases.

sistance maximum. Such behavior has been discussed in more detail in Refs. 23–26. The minimum which is observed in $\Delta\rho$ above about 10 K is due to the onset of a large positive contribution in the resistivity from deviations from Matthiessen's rule which overshadows the weakly decreasing Kondo contribution. The second maximum occurring aroung 50–70 K is due in part to deviations from Matthiessen's rule, but also to the breakup of magnetic clusters of impurities. As the clusters break up with increasing temperature the impurity resistivity decreases again. This behavior will be discussed more fully in Sec. IV. The data shown in Figs. 1–4 match nicely to previously published resistivity data for lower-concentration alloys.[23–26]

The low-temperature initial resistivity behavior was examined in more detail by plotting the data against various simple powers of T, and Fig. 5 shows some typical plots of $\Delta\rho$ versus $T^{3/2}$ for *Ag*Mn. It was found that down to the lowest temperature of measurement of 1.5 K (or, for certain *Au*Mn and *Au*Fe alloys, 0.5 K) a best fit to the data could be obtained with a simple expression: $\Delta\rho(T) = \Delta\rho_0(c) + A(c)\,T^{3/2}$. At the higher temperatures, the $\Delta\rho(T)$ behavior changes to a linear-in-T dependence; however, for some of the very lowest concentrations the 1.5-K minimum measuring temperature was unable to detect a clear $T^{3/2}$ region. Our data could of course be fitted to a more complicated polynominal, but above 1.5 K, a fit to a simple T^2 law was not suitable, except for the two highest concentrations of *Au*Cr (7.9 and 10.6 at.%), where there was a T^2 dependence at the lowest temperatures which was followed by a well-defined $T^{3/2}$ region, as is illustrated in Fig. 6. Additional examples of such $T^{3/2}$ plots were given for the other alloy systems, *Au*Fe, *Au*Mn, and *Cu*Mn, in our previous work.[1,18–20] Tables I–IV show that the temperature range of this $T^{3/2}$ dependence, T_1, increases with concentration. It is particularly large for the *Au*Cr alloys, roughly comparable to that for *Au*Fe, and is very small for *Au*Mn and especially *Ag*Mn. In this respect *Cu*Mn represents an intermediate situation between *Au*Cr and *Au*Fe on the one hand and *Au*Mn and *Ag*Mn on the other. However, despite these large differences in the range of the $T^{3/2}$ dependence between the various systems, it can be seen from Tables I–IV that there is much

TABLE I. Summary of resistivity data for *Au*Cr.

*Au*Cr concentration (at.% Cr)	0.9	1.5	3.3	4.9	7.9	10.6
$\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	4.319	5.768	13.363	18.945	28.955	38.535
$\Delta\rho$(4.2 K) ($\mu\Omega$ cm)	4.386	5.843	13.423	18.999	28.993	38.564
$\Delta\rho(T_\mu)$ ($\mu\Omega$ cm)	4.668	6.275	14.57	20.645	31.450	41.90
T_μ (K)	35	47	85	100	150	185
$\Delta\rho$(300 K) ($\mu\Omega$ cm)	4.116	5.473	13.60	19.61	30.65	41.2
$\Delta\rho$(4.2 K) $-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.067	0.075	0.060	0.054	0.038	0.029
$\Delta\rho(T_\mu)-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.349	0.507	1.207	1.700	2.495	3.365
$\Delta\rho(T_\mu)-\Delta\rho$(300 K) ($\mu\Omega$ cm)	0.552	0.802	0.97	1.035	0.800	0.700
$T_m(d\rho/dT \rightarrow \max)$ (K)	2±1	3±1	12±2	14±3	40±5[a]	45±5[a]
T_0[b] (K)	14	22	35	50	78	100
A [(nΩ cm)/K$^{3/2}$]	10.4	11.5	8.5	7.6	6.7	6.3
T_1(Range of $T^{3/2}$) (K)	3.2	3.7	5.8	9.7	20	21
T_1/T_0	0.23	0.17	0.17	0.19	0.26	0.21

[a] Upper end of the shoulder which has developed in $d[\Delta\rho(T)]/dT$.

[b] T_0 was determined from the sharp peaks in the low-field ac susceptibility. See Refs. 4, 9, and 11.

less variation when they are normalized with respect to the freezing temperatures: T_1/T_0; i.e., most of the values lie between 0.15 and 0.20. Similar values were obtained in *Au*Fe for concentrations less than 2 at.%.[1] Our analysis further shows that the coefficient of the $T^{3/2}$ temperature dependence, A, decreases very slowly with the concentration throughout this regime. Figure 7 illustrates this weak concentration dependence for all five alloy systems. The straight lines show that A is proportional to $-\ln c$, although rather strong departures away from this dependence seem to be observed for the lowest concentrations of *Cu*Mn and *Ag*Mn. An alternative slowly varying function, such as $c^{-1/5}$ for *Au*Fe and *Au*Cr and $c^{-1/2}$ for the three Mn systems, could equally well describe these data. The values obtained for A at for example $c = 5$-at.% impurity are, respectively, 7.5, 13, 15, and 12 nΩcm/K$^{3/2}$ for *Au*Cr, *Au*Mn, *Ag*Mn, and *Cu*Mn, which can be compared to 5.5 for *Au*Fe. These magnitudes suggest that there is a correlation between a long range over which the $T^{3/2}$ dependence is observed and the small value of the coefficient A. An additional correlation exists between a large magnetic scattering, as exemplified by large values of $\Delta\rho_0/c$ or $\Delta\rho(T_\mu) - \Delta\rho_0$, and the small magnitude of the $T^{3/2}$ coefficient A.

In a manner similar to I we have computer differentiated $\Delta\rho(T)$ to obtain the temperature coefficient of the resistivity $d[\Delta\rho(T)]/dT$. This method has been used quite extensively[27] on a wide variety of systems as a way of determining and characterizing the magnetic ordering. Various theories[28] support the experimental observation that the ordering temperature is distinguished by a divergence or sharp maximum in $d\rho/dT$. In Figs 8 and 9 we present such plots for the *Ag*Mn and *Cu*Mn alloy systems. Again, as with $\Delta\rho(T)$, quite similar behavior exists among these five systems. Namely, for sufficient magnetic concentration a low-temperature maximum appears

TABLE II. Summary of resistivity data for *Au*Mn.

*Au*Mn concentration (at. % Mn)	0.5[a]	1.5[a]	2.8	4.6	7.7	11.8
$\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	1.245	3.663	6.318	10.654	17.082	22.868
$\Delta\rho$(4.2 K) ($\mu\Omega$ cm)	1.284	3.743	6.407	10.736	17.148	22.923
$\Delta\rho(T_\mu)$ ($\mu\Omega$ cm)	1.324	3.950	6.745	11.485	18.630	25.470
T_μ (K)	60[b]	75	85	105	125	150
$\Delta\rho$(300 K) ($\mu\Omega$ cm)	1.265	3.812	6.594	11.245	18.283	25.090
$\Delta\rho$(4.2 K) $-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.039	0.080	0.089	0.082	0.066	0.055
$\Delta\rho(T_\mu)-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.074	0.290	0.425	0.825	1.550	2.600
$\Delta\rho(T_\mu)-\Delta\rho$(300 K) ($\mu\Omega$ cm)	0.059	0.138	0.151	0.240	0.347	0.380
$T_m(d\rho/dT \rightarrow \max)$ (K)	<0.5	1.5 ± 0.5	2.0 ± 0.5	3.0 ± 1.0	28 ± 2[c]	40 ± 8[c]
T_0[d] (K)	3.5	7.0	11	16.5	25	34
A [(nΩ cm)/K$^{3/2}$]	···	20.5	16.3	13.2	10.0	8.0
T_1(range of $T^{3/2}$) (K)	<0.5	1.45	2.7	3.2	3.6	4.7
T_1/T_0	···	0.21	0.24	0.19	0.14	0.14

[a] Measured down to 0.5 K.
[b] High-temperature maximum.
[c] Upper end of the shoulder which has developed in $d[\Delta\rho(T)]/dT$.
[d] T_0 was determined from the sharp peaks in the low-field ac susceptibility. See Refs. 4, 9, and 11.

in $d(\Delta\rho)/dT$, followed by a higher temperature falloff which gradually becomes negative, reflecting the maximum in $\Delta\rho(T)$. For the highest concentration, a broad shoulder displaces the maximum in $d(\Delta\rho)/dT$. As illustrated in Tables I–IV, there is *no* agreement, as was previously found for certain *Au*Fe concentrations, between the temperatures at which the maximum in $d(\Delta\rho)/dT$ occurs, T_m, and the spin-glass freezing temperature T_0. At best, some correlation exists with the upper shoulder region and T_0. For the four systems presently under study T_0 is always greater than T_m. In the case of *Au*Fe, $T_0 \simeq T_m$, but only for $c \gtrsim 3$ at.%. Therefore, in general, the temperature derivative of the magnetic resistivity does not directly characterize the freezing of the magnetic moments, as it would for a long-range periodic type of magnetic order.

IV. DISCUSSION

We will discuss the resistivity results in three parts. These are (a) the initial temperature and concentration dependence of $\Delta\rho$, (b) the behavior of $\Delta\rho$ and $d(\Delta\rho)/dT$ near the freezing temperature T_0, and (c) the high-temperature resistivity.

A. Initial temperature dependence

For all five systems presently under consideration, a substantial residual resistivity per at.% of impurity, $\Delta\rho_0/c$, exists. This is largest for *Au*Fe, followed by *Au*Cr, and then the three Mn doped systems. Such a relationship is consistent with resonant scattering at $T=0$ for a magnetic (spin-split) virtual bound state of $3d$ impurities.[29] Surprisingly, the magnitude of $\Delta\rho_0/c$ remains constant in each system for values of c up to $\simeq 5$ at.%—well

TABLE III. Summary of resistivity data for AgMn.

AgMn concentration (at. % Mn)	0.6	1.1	3.0	5.4	5.9	9.7
$\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.878	1.618	4.525	8.152	8.877	14.637
$\Delta\rho$(4.2 K) ($\mu\Omega$ cm)	0.927	1.689	4.621	8.249	8.973	14.714
$\Delta\rho(T_\mu)$ ($\mu\Omega$ cm)	0.960	1.782	5.009	9.080	9.904	16.374
T_μ (K)	47[a]	43	64	90	95	130
$\Delta\rho$(300 K) ($\mu\Omega$ cm)	0.854	1.622	4.735	8.683	9.413	15.750
$\Delta\rho$(4.2 K) $-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.049	0.072	0.096	0.097	0.096	0.077
$\Delta\rho(T_\mu)-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.082	0.164	0.484	0.928	1.027	1.737
$\Delta\rho(T_\mu)-\Delta\rho$(300 K) ($\mu\Omega$ cm)	0.106	0.160	0.274	0.397	0.491	0.634
$T_m(d\rho/dT\rightarrow\max)$ (K)	<1.5	<1.5	3.5 ± 0.5	4.5 ± 1.0	5.0 ± 0.5	6.5 ± 1.0
T_0[b] (K)	3.4	5.5	12	19	20.5	30
A [(nΩ cm)/K$^{3/2}$]	...	...	16.5	15.1	14.4	11.3
T_1(range of $T^{3/2}$) (K)	<1.5	<1.5	2.5	3.25	3.50	4.25
T_1/T_0	...	...	0.21	0.17	0.17	0.14

[a] High-temperature maximum.

[b] T_0 was determined from the sharp peaks in the low-field ac susceptibility. See Refs. 4, 9, and 11.

beyond the dilute limit. Thus, another contribution to $\Delta\rho_0$ might be expected from the randomly frozen spin glass. This would be a static type of magnetic disorder or defect, which for $T \ll T_0$, gives a temperature-independent potentiallike scattering and is in addition to the nonmagnetic types of defect or impurity scatterings. A separation of the magnetic residual resistivity might be accomplished by magnetoresistance measurements on these systems at low temperatures.

In this paper and in our previous work, it was emphasized that at low temperatures, down to 1.5 K (or in certain cases 0.5 K), the impurity resistivity had a well-defined $T^{3/2}$ initial temperature dependence with a coefficient which slowly decreased with increasing concentration. At higher temperatures, the $T^{3/2}$ function gradually deviates towards an approximately linear dependence around T_0, and slower temperature variations as $T > T_0$. We believe that this $T^{3/2}$ region has a physical significance rather than representing a slow transition between a T^2 and a T region. Similar $T^{3/2} \rightarrow T$ low-temperature dependences, but with a $T^{3/2}$ coefficient which decreases more strongly with concentration, have been observed in the dilute inhomogeneous ferromagnetic systems PdFe, PdCo, and PdMn.[30] This behavior has been interpreted[31] as due to a loss of momentum conservation in the electron-magnon interaction for an impure metal.[32] While a similar lack of translational invariance is present in the spin-glass case, no well-defined magnon modes would be expected.

A more appropriate treatment of the low-temperature resistive behavior of the spin glasses is the spin-diffusion theory of Rivier and Adkins[8] (RA), who ascribe the low-temperature resistive behavior to long-wavelength elementary excitations which are diffusive in character. These excita-

TABLE IV. Summary of resistivity data for *Cu*Mn.

*Cu*Mn concentration (at. % Mn)	0.7	1.6	2.7	4.5	6.3	9.7
$\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	2.176	4.733	7.747	12.009	16.452	24.222
$\Delta\rho$(4.2 K) ($\mu\Omega$ cm)	2.246	4.820	7.837	12.095	16.526	24.276
$\Delta\rho(T_\mu)$ ($\mu\Omega$ cm)	2.349	5.113	8.394	13.093	18.240	27.427
T_μ (K)	17	45	70	100	144	190
$\Delta\rho$(300 K) ($\mu\Omega$ cm)	2.088	4.735	7.866	12.481	17.713	27.040
$\Delta\rho$(4.2 K) $-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.071	0.088	0.090	0.086	0.074	0.054
$\Delta\rho(T_\mu)-\Delta\rho$(1.5 K) ($\mu\Omega$ cm)	0.174	0.380	0.647	1.084	1.788	3.205
$\Delta\rho(T_\mu)-\Delta\rho$(300 K) ($\mu\Omega$ cm)	0.261	0.378	0.528	0.612	0.527	0.387
$T_m(d\rho/dT \rightarrow \max)$ (K)	2.0 ± 0.5	3.5 ± 0.5	4.5 ± 0.5	5.0 ± 0.5	10 ± 1[a]	50 ± 5[a]
T_0[b] (K)	8	13	18	27	33	44
A [(nΩ cm)/K$^{3/2}$]	•••	15	14	12.6	10.7	7.7
T_1(range of $T^{3/2}$) (K)	< 1.5	2.5	3.2	4.0	5.0	7.0
T_1/T_0	•••	0.10	0.18	0.15	0.15	0.16

[a] Upper end of the shoulder which has developed in $d[\Delta\rho(T)]/dT$.

[b] T_0 was determined from the sharp peaks in the low-field ac susceptibility. See Refs. 4, 9, and 11.

tions are highly damped, noncoherent (independent), localized spin fluctuations which scatter the conduction electrons. Following the general conductivity equation, RA evaluate the relaxation time within the multiple-scattering approximation. Their calculation of the resistivity proceeds along lines similar to that previously used by Rivier and Zlatić.[33] For purely diffusive modes, the low-temperature resistivity becomes $\Delta\rho(T, c) = c\rho(0) + A(c)\,T^{3/2}$. Here the residual resistivity $c\rho(0)$ is due to resonant scattering from a spin-split virtual bound state and

$$A(c) = \rho(\infty)\, D^2 \left(\frac{JS}{\Gamma}\right)^2 \frac{a^3}{4}\left(\frac{\pi}{2}\right)^{1/2}\left(\frac{k_B}{\Lambda a^{-2}}\right)^{3/2} .$$

$\rho(\infty)$ is the unitarity scattering limit $[=\frac{1}{2}\, c(m/\hbar e^2) \times \Gamma(n/\Omega)^{-1}]$, with Γ the conduction bandwidth and n/Ω the electron concentration. Other parameters in the expression for $A(c)$ include D, the virtual bound states's ± displacement from Fermi level to width parameter; J, the spin (S)-spin coupling constant; k_B, the Boltzmann constant; a, the lattice spacing; and Λ, a diffusion constant. The concentration dependence in $A(c)$ comes from $\rho(\infty) \sim c$ and $\Lambda \sim cf(l(c))$, where f is a complicated but slowly varying function of the electronic mean free path $l(c)$. Thus, $A(c) \sim 1/\{c[f(l(c))]^3\}^{1/2}$. According to RA, above a certain temperature T_1, directly proportional to Λ, the $T^{3/2}$ resistive behavior is no longer valid and $\rho(T)$ gradually flattens out with a corresponding point of inflection at T_m, where $d\rho/dT$ exhibits a maximum. There is no unique physical connection between T_m in this model and the freezing temperature T_0 or process. At very high temperatures the unitarity limit is reached. The calculations of the extended temperature behavior yield certain relationships [see

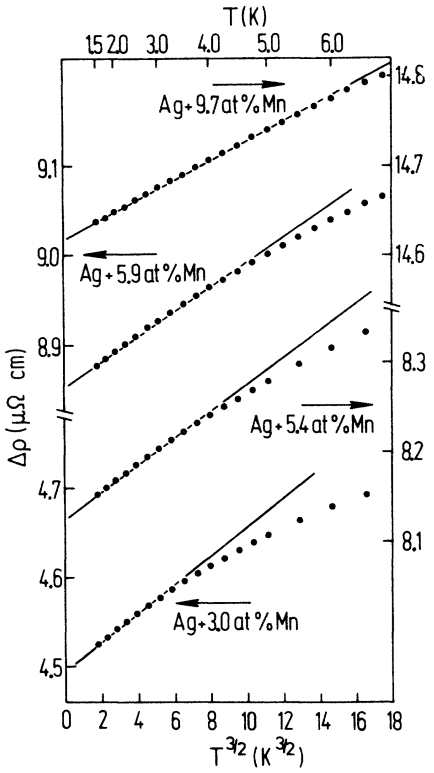

FIG. 5. $\Delta\rho$ ($\mu\Omega$ cm) plotted against $T^{3/2}$ ($K^{3/2}$) for some AgMn alloys.

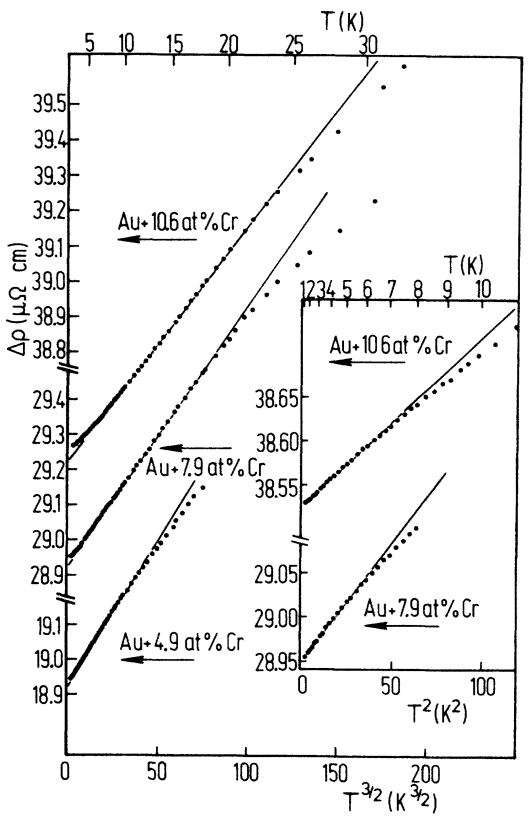

FIG. 6. $\Delta\rho$ ($\mu\Omega$ cm) plotted against $T^{3/2}$ ($K^{3/2}$) for the three highest concentrations of AuCr alloys. The insert shows the initial values of $\Delta\rho$ plotted against T^2 for the 7.9- and 10.6-at.% Cr alloys. Note the change in scale for $\Delta\rho$ in the $T^{3/2}$ and T^2 plots.

RA, Eqs. (23)–(25)] between T_1, T_m, $\rho(\infty)D$, and A which are mainly independent of the alloy system and its concentration.

The general features of this theory are in good qualitative agreement with the experimental results for all five systems, namely, (a) a large residual resistivity from the spin-split virtual bound state, (b) a $T^{3/2}$ initial temperature dependence whose coefficient slowly decreases with increasing concentration, (c) a large range of $T^{3/2}$ behavior ($T_1 \sim \Lambda$) causing a small magnitude of the coefficient $A \sim (1/\Lambda)^{3/2}$, and (d) a point of inflection in $\Delta\rho(T)$ at a characteristic T_m followed by a much slower temperature variation at higher temperatures. A more detailed quantitative comparison is carried out in RA for a Au + 1-at.%-Fe alloy. In addition, we have performed a quantitative test of the "canonical" relationship between T_1, T_m, $\rho(\infty)D$ [corresponding to our $\Delta\rho(T_\mu) - \Delta\rho(1.5\ \mathrm{K})$], and A using the data given in Tables I–IV, and find reasonable agreement among the various alloys despite the stringent requirements of these relationships and some large experimental uncertainties.

The low-temperature $T^{3/2}$ dependence in the RA theory is very sensitive to the availability of very-long-wavelength diffusion modes, i.e., those with a very low wave number q. If these modes are no longer operative, i.e., a $q_{\min} \neq 0$, owing to spin relaxation with nonmagnetic impurities, or surface, defect, or grain boundary scattering, then

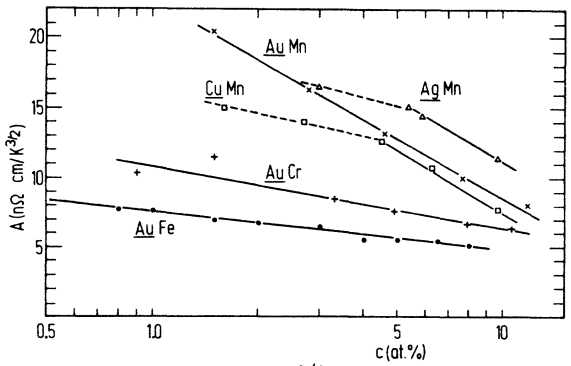

FIG. 7. Coefficient of the $T^{3/2}$ dependence A (nΩ cm/$K^{3/2}$) plotted against $\log_{10}c$ for the AuCr, AuMn, AgMn, and CuMn alloys. Data for AuFe alloys taken from I are also shown for comparison.

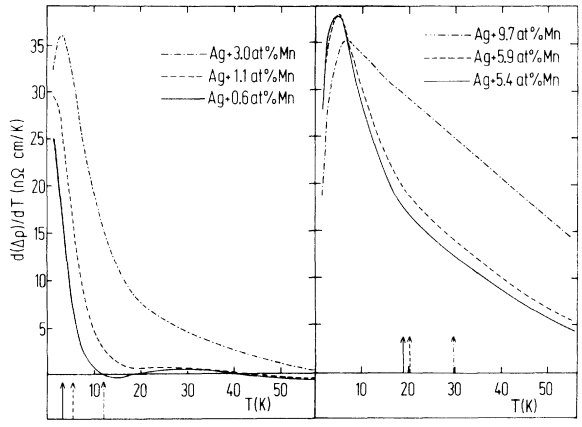

FIG. 8. Temperature dependence of the temperature derivative of the impurity resistivity $d(\Delta\rho)/dT$ (nΩ cm/K) for the AgMn alloys. The arrows represent T_0 as determined from the susceptibility measurements; see Refs. 4 and 11.

a T^2 dependence can supplant the $T^{3/2}$ behavior at the lowest temperature. This may account for the T^2 temperature dependence which has been observed for $T \lesssim 0.3$ K on some lower concentrations CuMn, AuFe, and AuMn alloys,[34] as well as in the two most concentrated AuCr samples of the present work; see Fig. 6. Furthermore, this T^2 occurrence should be much more sensitive to changes in the nonmagnetic potential scattering, as would be produced by different heat treatments. To date such effects have not been tested experimentally.

We might now mention two shortcomings of this experimental-theoretical comparison. Our measurements show a correlation between a large $\Delta\rho_0$ and large amount of magnetic scattering between $T \simeq 0$ and T_μ, e.g., $[\Delta\rho(T_\mu) - \Delta\rho_0]/c$, while the theory suggests the opposite. Also the high-temperature theoretical treatment has not taken into account possible kinematic interactions or Kondo-like effects, which would cause the maximum in $\Delta\rho$ and the negative $\Delta\rho$ slope as room temperature is reached.

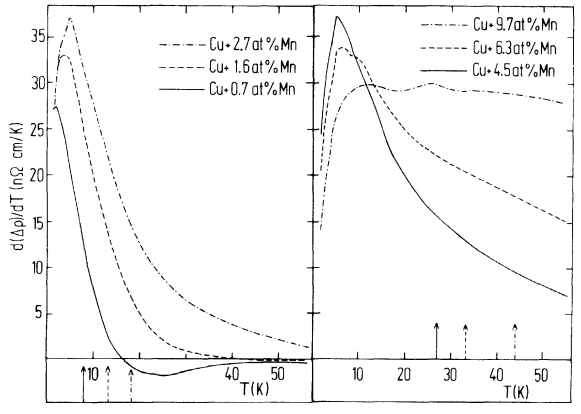

FIG. 9. Temperature dependence of the temperature derivative of the impurity resistivity $d(\Delta\rho)/dT$ (nΩ cm/K) for the CuMn alloys. The arrows represent T_0 as determined from the susceptibility measurements; see Refs. 4 and 11.

B. Behavior of $\Delta\rho$ and $d(\Delta\rho)/dT$ near T_0

There exists no definite indication from $\Delta\rho$ or $d(\Delta\rho)/dT$ of the spin-glass freezing temperature T_0, which is clearly observed in such measurements as the susceptibility, Mössbauer effect, etc.[4,35] Spanning T_0, $\Delta\rho$ possesses a rapidly rising behavior, but the maximum value of its slope $[d(\Delta\rho)/dT - \text{max.}]$ lies, in general, at a temperature T_m considerably below T_0. Figures 8 and 9 show T_0 to occur in a region near the maximum negative slope of $d(\Delta\rho)/dT$, i.e., closer to the minimum in $d^2(\Delta\rho)/dT^2$. The exceptions to this prevalent behavior are the AuFe system, where $T_m \simeq T_0$ for $c \gtrsim 3$-at.% Fe, and the highest concentration of the other systems where a shoulder develops in $d(\Delta\rho)/dT$ at $\simeq T_0$. This behavior leads us to speculate about the nature of the freezing process in the following manner. Our resistivity studies are consistent with the absence of long-range cooperative magnetic order in the freezing of a spin glass or mictomagnet, in so much as any type of macroscopically periodic spin structure occurring at a well-defined temperature should be reflected in $\Delta\rho(T)$, or especially in $d(\Delta\rho)/dT$. Nevertheless, considerable short-range or local, directly coupled, magnetic ordering (clustering) appears and grows at temperatures well above T_0—the Sec. IV C will more fully discuss the high-temperature behavior. In the temperature region around T_0, the significant drop in $\Delta\rho(T)$ as the temperature is lowered indicates a large decrease in the dynamical spin-disorder scattering. Although this effect of the spin freezing is clearly seen in the resistivity, it does not take place at a certain, distinct, temperature, but is smeared out over a wide temperature interval. This probably has to do with the particular averaging process involved with a resistance measurement. To understand this a little better, we offer an oversimplified model. Let us divide our alloy into two types of spins: those already in clusters strongly coupled to their neighbors, and those, making up the spin-glass matrix, relatively isolated, weakly interacting via the long-range Ruderman-Kittel-Kasuya-Yosida mechanism. As the temperature is lowered, the clusters expand

in both size and number, resulting in a decrease in $\Delta\rho(T)$—less spin disorder scattering. At $T = T_0$, the spin-glass matrix locks into random orientations, the theoretical descriptions of which are given by Edwards and Anderson.[14] This is followed by the larger clusters becoming blocked, owing to a slight shift in their energy barrier potentials caused by the sudden freezing of the spin matrix. However, here there is a large distribution of barrier heights within this otherwise conventional superparamagnet picture. The barrier heights are related to an internal field H which is determined from the probability distribution $P(H, T)$.[36] So only a small group of the net spins of the alloy cooperatively take part in the freezing process. For an indirect measurement such as the resistivity, thermopower,[37] or the specific heat,[38] where strong, additional, nonmagnetic contributions are present, the effects of the freezing process possess insufficient strength to clearly show up within the available experimental sensitivity. In contrast, a direct magnetic measurement, e.g., the susceptibility or the Mössbauer effect, does have the intrinsic sensitivity to detect this relatively weak freezing behavior at T_0.

If the magnetic clusters are sufficiently large, i.e., average radius r_0 greater than the electronic mean free path l, then a better chance should exist to detect the cluster freezing in $d(\Delta\rho)/dT$. This would be the case for moderately concentrated AuFe alloys, which have the smallest mean free path of the systems presently under consideration and large ferromagnetic clusters.[39] The above criterion, $r_0 > l$, would also hold for the highest concentrations ($\simeq 10$ at.%) of the other systems, where the shoulder in $d(\Delta\rho)/dT$ develops, and can roughly be correlated with T_0. It should be noted here that the details of the spin freezing and spin structure are complicated and not completely understood. Furthermore, there is, at present, little theoretical understanding of how such effects would manifest themselves in a transport property.

C. High-temperature resistivity

The analysis of the data at temperatures where the phonon contribution is important is made difficult by uncertainties arising from at least three factors. These are (a) the modifications of the host lattice by the addition of several at.% of impurities, (b) the effects of deviations from Matthiessen's rule, and (c) the thermal expansion of the alloys. The possible effects of these three factors will be discussed later in this section. Nevertheless, because of them, it must be emphasized that the following arguments should be regarded as somewhat speculative.

The resistance maxima of the four systems presented in this paper are clearly defined, much more so than the maxima of AuFe, which are less resolvable owing to their occurrence at temperatures near or greater than 300 K. As the concentration of impurities increases, the resistance maximum shifts to higher temperatures and also becomes broader. At temperatures above T_μ, the alloys of all four systems exhibit a well-defined region where the impurity resistivity is decreasing linearly with increasing T. Examination of Figs. 1–4 show that this linear dependence frequently exists for a range of $\simeq 150$ K. In some of the less-concentrated alloys, $c \simeq 1$ at.%, $\Delta\rho(T)$ varies slower than T for temperatures above about 200 K and tends towards a $\log_{10}T$ dependence. The available data[40] on these systems, up to temperatures of around 1000 K, show that at these very high temperatures the impurity resistivity approximately decreases like $\log_{10}T$. For temperatures slightly greater than T_μ, $\Delta\rho$ is decreasing faster than T, and there are regions where a $1 - T^2$ fit to the data would be appropriate. We, therefore, believe that with increasing temperature the high-temperature decrease in the impurity resistivity is describable in terms of a succession of $1 - T^2$, $1 - T$, and $-\log_{10}T$ dependences. Similar variations are predicted[41] and observed[42,43] for spin localized-fluctuation systems such as AlMn and AuV.

In our previous papers we have suggested that the resistivity maximum at T_μ approximates the temperature at which magnetic clusters begin to occur. The various systems exhibit differences among themselves, for example, in the magnitude of T_μ, which indicates the strength of the cluster formation, or in the sharpness of the maximum, which is particularly distinct for the AuCr system. However, there is a definite similarity among these systems in the over-all behavior. A recent theoretical model[44] utilizing intracluster excitations has been proposed to treat the high-temperature resistance behavior for CuNi alloys which also have a ρ maximum and a high-T falloff. Although this model has not yet been extended to include the spin glasses, nevertheless, the basic cluster dynamics should, in general, produce parallel resistivity effects.

In the above discussion no account has been taken of the three factors mentioned at the beginning of this subsection which could substantially alter the observed temperature dependence of $\Delta\rho$ in a manner difficult to assess. We will now briefly discuss the possible influence of each factor.

The addition of 3d impurities to the noble metals

is expected to produce a change in the phonon spectrum, since there are marked differences between the solvent and solute atoms in their atomic weights, structures, Debye temperatures, and room-temperature resistivities.[45] Such perturbations of the host phonon spectrum have hardly been investigated, and therefore, the simple subtraction of the resistivity of the noble metal from that of the alloy must be regarded as an approximation, albeit the best available to us at the present time.

Much of the extensive studies of deviations from Matthiessen's rule (DMR) has been summarized in two review articles by Bass[46] and by Cimberle *et al.*[47] These show that as the phonon resistivity becomes large, measurable effects from DMR can occur and have a variety of temperature and concentration dependences. Numerous theories have been advanced to explain these observations and among them the "two-band" model[48] has been extensively used for impurities in the noble metals. In particular, this model was employed by Whall *et al.*[49] in their investigations of dilute alloys ($\lesssim 0.1$ at.%) of $3d$ transition metals in *Au*. Here they observed significant effects due to DMR which at low temperatures were proportional to the phonon resistivity. The two-band model was further used in an analysis of the impurity resistivity of *Au*V alloys,[43] where it was concluded that DMR were seriously modifying the details of the observed temperature dependence. An attempt to include DMR based upon this approach was made in I. By extrapolating to higher concentrations the results of Ref. 49, it was estimated that DMR became important above $\simeq 50$ K, and, in the case of 1- and 2-at.% Fe alloys, could account for almost the entire change in $\Delta\rho$ between 50 and 300 K. In addition, it was believed that when DMR corrections were included, there would be a stronger falloff in $\Delta\rho$ at higher temperatures than that shown in Fig. 2 of Ref. 1, and also that the maximum in $\Delta\rho$ would be shifted to lower temperatures. For higher Fe concentrations we estimated the DMR to become progressively less important owing to increasing magnetic contributions. In general, for larger nonmagnetic impurity concentrations,[48] the DMR constitute a much smaller percent of the residual resistivity and possess a simple step-function-like temperature dependence. We expect the effects of DMR for the four systems discussed in this paper to be similar to those suggested for *Au*Fe in I because of the rather close nature of the Cr, Mn, and Fe impurities. Furthermore, we believe that DMR are not responsible for the overall features of our experimental observations, although they might strongly influence the details of the temperature and concentration dependences above about 50 K. However, until a systematic study of DMR is completed for concentrated, nonmagnetic, alloys and combined with some theoretical model, a more accurate evaluation of the high-temperature resistive behavior of the spin glasses is not possible.

Additional uncertainties in a large-temperature-range resistivity evaluation come from thermal-expansion effects, which can alter the sample geometry and the atomic volume. Since the coefficients of thermal expansion are relatively constant for the three noble metals below liquid-nitrogen temperatures, our low-temperature data are completely unaffected by these effects. Above $\simeq 100$ K we could include a temperature-dependent sample-geometry factor, which would lead to corrections on average of about 10% of the net resistivity: $\Delta\rho(100\text{ K}) - \Delta\rho(300\text{ K})$. However, as considered above, we are unable to perform a quantitative evaluation of these high-temperature data owing to phonon spectrum modifications and DMR. High-pressure measurements[50] on moderately concentrated spin-glass alloys show very little pressure dependence for $\Delta\rho(T)$. This indicates that changes in the atomic volume which can alter the effective s-d exchange interaction, as have been observed for Kondo[51] and spin-fluctuation[52] systems, play no role here.

ACKNOWLEDGMENTS

The authors have benefited from useful discussions with K. Adkins, V. Cannella, H. Claus, J. R. Cooper, K. H. Fischer, U. Larsen, N. Rivier, and J. S. Schilling. We acknowledge the hospitality shown to us by the Metal Physics Group, Imperial College, London, during our numerous visits. Finally we thank S. Britz and S. Hedwig for assistance with the computer analysis.

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