Electrical-resistivity studies on *Au*FeCr

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Electrical-resistivity(ρ) measurements are reported for a series of $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys ($0 \leq x \leq 0.8$) in the temperature range 4.2 K–300 K. The system emerges into a cluster glass or spin glass for $x > 0.1$, with the end alloys $Au_{82}Fe_{18}$ and $Au_{82}Cr_{18}$ being reentrant and antiferromagnetic, respectively. The alloys with $x \geq 0.6$ show pronounced resistivity maxima. The "impurity" resistivity $\Delta\rho$ shows a $T^{3/2}$ dependence. The saturation of resistivity for $x \geq 0.4$ is discussed in terms of the mean-free-path damping of the Ruderman-Kittel-Kasuya-Yosida interaction. However, a quantitative fit to a theory by Larsen does not hold for this system.

I. INTRODUCTION

There have been extensive studies on the problem of localized moments in dilute magnetic alloys. Several systems have emerged which exhibit Kondo, spin-glass, and reentrant-spin-glass (RSG) behavior depending on the concentration of the magnetic impurity in the nonmagnetic host. In canonical spin glasses such as *Au*Fe, *Au*Mn, *Cu*Mn, *Ag*Mn, *Au*Cr, etc., there have been detailed studies to determine their magnetic and electrical transport behavior.[1–3] In the RSG regime where a double magnetic transition is seen, the situation is still quite complicated and there still persists strong controversy about the origin of the RSG transition which evolves from an inhomogeneous ferromagnetic phase. In *Au*Fe and *Au*Cr systems, the experimentally determined magnetic phase diagrams show a long-range ferromagnetic and antiferromagnetic behavior, respectively, for Fe and Cr concentrations above 15 at.%.[4,5] We have therefore prepared $Au_{82}(Fe_{1-x}Cr_x)_{18}$ with $0 \leq x \leq 0.8$ to study how the RSG behavior of $Au_{82}Fe_{18}$ is modified as a result of Cr substitution.

Systematic studies of electrical resistivity in the temperature range 0.5–300 K and varying impurity concentrations have been made by Ford and Mydosh in canonical spin-glass systems like *Au*Fe,[2] *Au*Cr, *Au*Mn, *Ag*Mn, and *Cu*Mn.[3] In all these systems, the impurity resistivity, $\Delta\rho$, has an initial $T^{3/2}$ dependence with a linear T dependence near the freezing temperature, T_f, followed by a resistance maximum at a higher temperature. The first derivative of impurity resistivity has a well-defined maximum but only in the case of *Au*Fe does it nearly coincide with T_f.

The low-temperature variation of resistivity has been theoretically explained in terms of the variation of the spin-glass order parameter by Seiden[6] and elementary excitations of a diffusive nature by Rivier and Adkins[7] and Fischer.[8] The resistivity maximum is linked to an interplay of spin-glass properties and the Kondo effect.[9,10] Detailed studies made on binary spin-glass alloys are available, more so in *Au*Fe where temperature, magnetic field, pressure, etc., have been varied. Larsen has comprehensively compiled data available on *Au*Fe in terms of characteristic temperatures like the spin-glass freezing temperature, T_f, temperature of maximum in resistivity, T_m, etc., and has tried to explain their observed dependences on concentration and electronic mean free path employing a theoretical calculation based on Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange interaction between the impurity spins.[11] In case of *Au*Fe, three distinct regimes in terms of Fe concentrations (c) emerge. Kondo behavior is observed for $c \leq 0.05$ at.%, while the spin-glass and inhomogeneous ferromagnetic regimes exist for 0.5 at.% $\leq c \leq 10$ at.% and $c \geq 15$ at.%, respectively. At the two extreme limits, there is a departure from the theoretical predictions for characteristic temperatures and concentration dependence. Incorporating the Kondo effect, some agreement with the RKKY calculation of the interaction strength is obtained while for the higher concentrations where nearest-neighbor direct-exchange interactions dominate, theories based on the percolation model can only give some qualitative interpretation.

Studies of ternary spin-glass alloys mainly involved studying effects of adding a nonmagnetic or magnetic impurity[12,13] to canonical spin-glass systems. Sherlekar *et al.*[14] have studied resistivity in a ternary spin-glass system *AuCu*Mn and have analyzed the resistivity maximum in terms of Larsen's theory.[9] A $T^{3/2}$ dependence of the impurity resistivity, $\Delta\rho$, obtained by subtracting out the resistivity of the host, is also observed in *AuCu*Mn. From their studies on *Ag*MnSn, *Au*FeCr, *AuCu*Mn, Vier and Schultz[13] claim that both mean free path and spin-orbit effects contribute to spin-glass freezing temperature. Larsen has however shown that the theory based on saturation of the RKKY interaction damping alone accounts for the observations made by Vier and Schultz in the case of *Ag*MnSn.[15] In the light of the Kaneyoshi model[16] which calculates the indirect-exchange interaction in disordered magnets using a one-electron Green function, Larsen shows that the RKKY interaction damping accounts for the observed spin-glass freezing temperature T_f even in ternary alloys AB_xC_y

where A is the nonmagnetic metal host, B is the dilute nonmagnetic species of concentration x, and C is a magnetic impurity of concentration y. In the case of ternary alloys with more than one magnetic impurity in a nonmagnetic host, the situation is more complex because of more than one type of magnetic interaction, especially if the magnetic impurity concentration is near the percolation threshold. This is the case in our alloys, $Au_{82}(Fe_{1-x}Cr_x)_{18}$, where the low-field ac susceptibility studies show that with increasing Cr concentration x, it changes into pure spin glass, presumably as a result of competing ferromagnetic and antiferromagnetic exchange interactions.[17]

II. EXPERIMENTAL DETAILS

The samples of $Au_{82}(Fe_{1-x}Cr_x)_{18}$ with $x = 0$, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.6, and 0.8 were prepared by arc melting the high purity constituents in an argon atmosphere. They were homogenized and annealed at 850 °C for 1 week. They were cold rolled to foils of thickness of around 100 μm from which rectangular strips were cut for measurement. These strips were annealed at 850 °C for 24 h and quenched in water (two of the alloys, $x = 0.15$ and 0.25 were quenched in liquid nitrogen) and stored in liquid nitrogen until measurements were made. X-ray-diffraction (XRD) measurements on these samples confirmed the existence of a single phase and the lattice constant determined from them varied from 4.03 Å for the $x = 0$ alloy to 4.05 Å for $x = 0.8$ while that of pure Au is 4.08 Å.

Electrical resistivity measurements were carried out using the standard dc four-probe technique in the temperature range 4.4–300 K. A calibrated Lakeshore silicon diode sensor was used to monitor the sample temperature which was controlled using a DRC-82C Lakeshore temperature controller. Current and voltage leads were spot-welded to the sample. All measurements were automated using IBM-PC to which instruments were coupled via IEEE-488 interface. The stability in measurements was better than 50 ppm.

III. RESULTS AND DISCUSSION

The magnetic contribution to the resistivity, $\Delta\rho(T)$, is obtained by subtracting the resistivity of the noble metal host (Au) at each temperature from the total resistivity. The plots of $\Delta\rho(T)$ as a function of temperature are shown in Figs. 1 and 2. For alloys with x= 0.05–0.4, $\Delta\rho(T)$ increases significantly with T followed by a slower increase up to room temperature. There appears a clear maximum in $\Delta\rho(T)$ for alloys with $x = 0.6$ and 0.8 as shown in Fig. 2. This maximum is more pronounced for the higher Cr concentration alloy ($x = 0.8$). Though the ac susceptibility data show a spin-glass-like sharp cusp for the $x = 0.4$ alloy at 57.5 K,[17] there is no evidence of a maximum in $\Delta\rho(T)$. Since $\Delta\rho(T)$ seems to be saturating as we approach 300 K, it is quite likely that the resistivity maximum in this alloy might occur above this temperature. The first derivative of resistivity, $d(\Delta\rho)/dT$, shows a maximum which does not yield any direct correlation with the spin-glass transition temperature, T_f, as had been the case in *Au*Fe alloys. The low-temperature resistivity data were computer fitted to the relation

$$\Delta\rho(T) = \Delta\rho(T) + AT^{3/2}$$

using standard IMSL (International Mathematical and Statistical Libraries) routines. The range of best fit was decided by the least value of chi square (χ^2) or χ_{RMSD} where

$$\chi^2 = \sum_{i=1}^{N} \frac{(\rho_i^{\mathrm{expt}} - \rho_i^{\mathrm{calc}})^2}{\rho_{\mathrm{mean}}^2},$$

$$\chi_{\mathrm{RMSD}} = \sqrt{\frac{\chi^2}{N}},$$

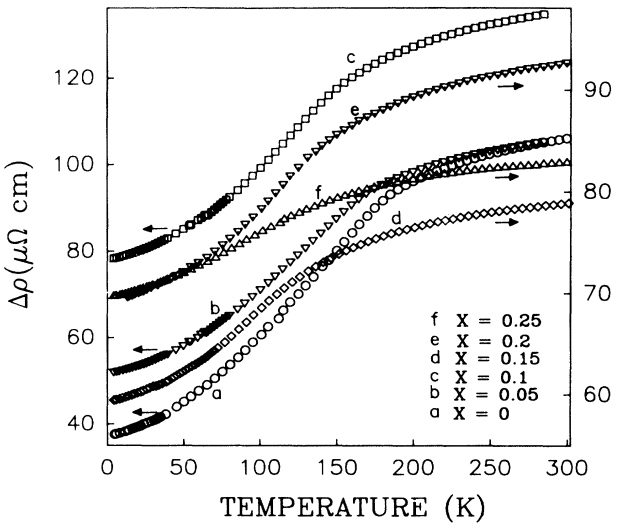

FIG. 1. Temperature (T) variation of $\Delta\rho(\mu\Omega\,\mathrm{cm})$ of $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys for $0 \leq x \leq 0.25$.

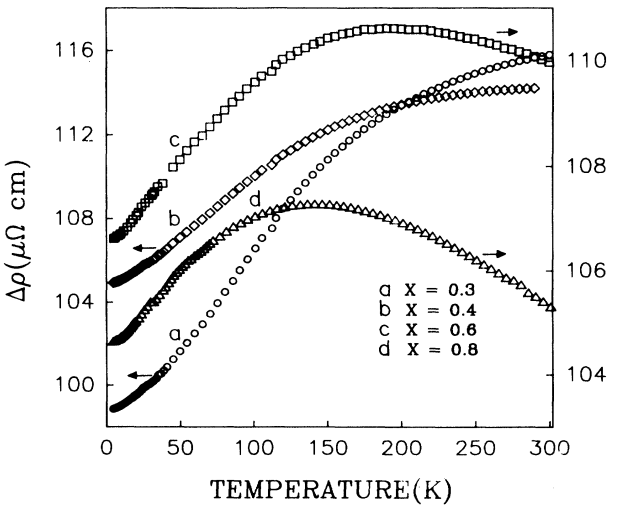

FIG. 2. Temperature (T) dependence of $\Delta\rho(\mu\Omega\,\mathrm{cm})$ of $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys for $0.3 \leq x \leq 0.8$.

TABLE I. Results of fitting $\Delta\rho$ to $\Delta\rho_0 + AT^{3/2}$. x is the Cr concentration in $Au_{82}(Fe_{1-x}Cr_x)_{18}$. T_1 indicates range of fit. Freezing temperature T_f and Curie temperature T_c (in the case of double transition) are from Ref. 17. χ_{RMSD} is as defined in text. Mean free path, λ at 4.4 K, is calculated from Eq. (1) using the free electron model.

x	$T_f(T_c)$ (K)	$\Delta\rho_0(\mu\Omega\,\mathrm{cm})$	A (nΩ cm/K$^{3/2}$)	T_1 (K)	χ_{RMSD} (10^{-4})	λ (Å)
0	65.1(150.7)	37.41	21.68	4.4–26	6.56	22.32
		36.22	24.25	65–135	10.9	
0.05	71.6(127.0)	51.74	17.76	4.4–45	4.32	16.19
		50.33	20.61	68–130	6.42	
0.1	69.4(109.5)	78.20	19.58	4.4–38	2.41	10.72
		75.76	23.52	68–134	10.1	
0.15	66.6	59.40	8.88	4.4–26	1.55	14.12
0.2	73.3	69.24	8.19	4.4–60	3.54	12.12
0.25	56.6	69.78	6.88	4.4–42	2.80	12.03
0.3	69.7	98.80	8.02	4.4–60	3.11	8.50
0.4	57.5	104.88	6.42	4.4–45	2.99	8.00
0.6		106.54	5.37	4.4–20	2.47	7.88
0.8		104.60	3.99	4.4–11	0.47	8.03

N is the number of data points, ρ_i^{expt} are experimentally measured values, ρ_i^{calc} are theoretically calculated values, and ρ_i^{mean} is mean of the experimental values.

The values of residual resistivity, $\Delta\rho_0(\mu\Omega\,\mathrm{cm})$, the coefficient of the $T^{3/2}$ term, A (nΩ cm/K$^{3/2}$), and the range of fit are given in Table I. In the case of $x = 0$, 0.05, and 0.1 where the χ_{ac} shows a distinct reentrant behavior, we find two distinctly different temperature ranges following the $T^{3/2}$ dependence, one below the RSG transition temperature and the other above it as can be seen in Fig. 3. It is interesting to note that the higher temperature $T^{3/2}$ fitting has the same temperature range for all the three alloys (65–135 K) though their Curie temperatures (T_c) are significantly different. This higher temperature $T^{3/2}$ dependence was apparently overlooked by Mydosh *et al.* in *Au*Fe alloys.[2] It clearly indicates that there is a different or additional magnetic scattering mechanism for resistivity in the ferromagnetic region. It is possible that the low-temperature dependence below the RSG transition is due to the diffusive nature of spin flip excitations while the higher-temperature dependence may be caused by incoherent electron-magnon scattering as proposed by Mills *et al.* for dilute ferromagnets.[18] The values of A for the reentrant alloys are weakly concentration dependent whereas there is a sudden drop when the concentration changes from reentrant to cluster glass, i.e., for $x \geq 0.15$. This is further followed by a similar weak concentration dependence for the other alloys ($0.2 \leq x \leq 0.8$). For alloys in the cluster glass regime, Mydosh *et al.* found that the $T^{3/2}$ dependence of $\Delta\rho$ exists up to near the freezing temperature.[2] This range drastically reduces in the pure spin-glass regime. A behavior similar to that of cluster glass is found in the present study for alloys with $0.2 < x < 0.4$ (Fig. 4).

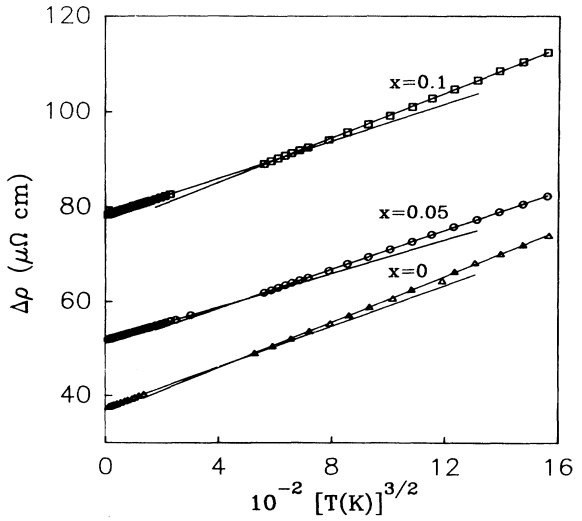

FIG. 3. $\Delta\rho$ plotted against $T^{3/2}(K^{3/2})(10^{-2})$ for the reentrant alloys. The solid lines are results of fit to $\Delta\rho_0 + AT^{3/2}$ (see text).

A significant feature of the present data is that the residual resistivity $\Delta\rho_0(T)$ increases from around 37 $\mu\Omega$ cm for the $x = 0$ alloy to nearly 100 $\mu\Omega$ cm for alloys with $x = 0.3$. It is nearly constant for alloys with $x \geq 0.3$ as shown in Fig. 5. This value of 100 $\mu\Omega$ cm seems to be high for these alloys which implies that the alloys with large chromium are highly disordered. Again, the value of residual resistivity for the end alloy $Au_{82}Cr_{18}$ (which shows long-range antiferrogmagnetic order at 235 K as observed by Nakai *et al.*[5] and corroborated by SQUID measurements made by us) falls to 81 $\mu\Omega$ cm, which is almost twice that of $Au_{82}Fe_{18}$ ($\simeq$37 $\mu\Omega$ cm). However, for alloys in the spin-glass region ($c \leq 10$ at. %), it is seen from the measurements of Mydosh *et al.* that the values of resistivity for the *Au*Cr alloys were less than those of the corresponding *Au*Fe alloys.[2,3] In the case of *Au*Fe alloys, the residual resistivity drops after $c > 12$ at. %.[2] From the present observations, it appears that in the case of *Au*Cr alloys, there is a monotonous increase in the resistivity values with increasing Cr concentration.

In Fig. 5, both $\Delta\rho_0$ and $(\Delta\rho_{HT}-\Delta\rho_0)$, have been shown as a function of x for $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys where $\Delta\rho_{HT}$ is the magnetic resistivity at room temperature. It is interesting to note that $(\Delta\rho_{HT}-\Delta\rho_0)$ falls off quite rapidly as x increases from 0 to 0.4 and then

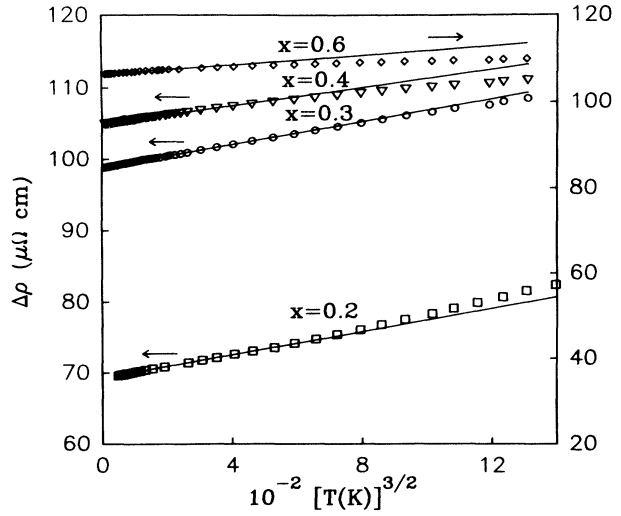

FIG. 4. $\Delta\rho$ vs $T^{3/2}(K^{3/2})(10^{-2})$ for $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys for Cr concentrations indicated.

decreases slowly. This indicates that the degree of ferromagnetic alignment decreases rapidly with increasing Cr concentration. In *Au*Fe alloys with Fe concentration above 12 at.%, $(\Delta\rho_{HT}-\Delta\rho_0)$ has been found to increase rapidly with Fe concentration showing a strong degree of ferromagnetic alignment of Fe spins with decreasing temperature.[19] The present results thus show that the presence of Cr in *Au*Fe rapidly decreases this tendency of ferromagnetic alignment or in other words, it causes anticlustering in the system.

Two of the alloys with Cr concentration $x = 0.15$ and 0.25 were quenched from 850 °C into liquid nitrogen unlike the other alloys that were quenched into water. A greater degree of disorder, hence an increased value of resistivity, was expected. However, it was observed that the values were much lower as is shown in Figs. 1 and 5.

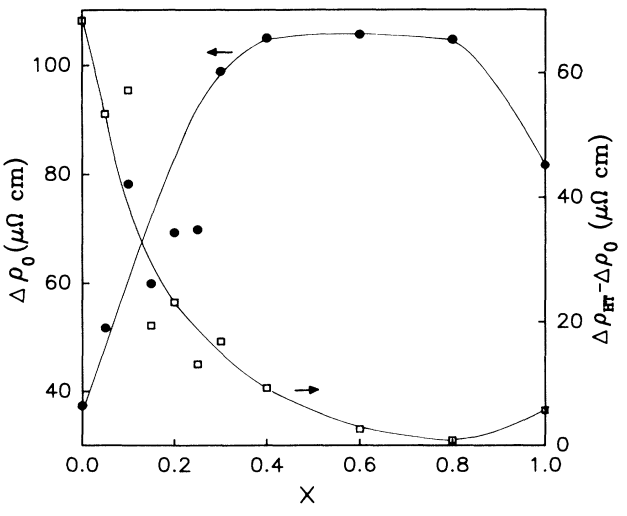

FIG. 5. Plot of $\Delta\rho_0$ and $(\Delta\rho_{HT}-\Delta\rho_0)$ (defined in text) vs Cr concentration x. The solid lines are a guide to the eye.

The values of the electronic mean free path λ were calculated from the expression

$$\rho\lambda = 0.84 \times 10^{-11}\ \Omega\,\mathrm{cm}^2, \tag{1}$$

which is based on a free electron model.[20] For $Au_{82}Fe_{18}$, λ is calculated to be 22 Å at 4.4 K and falls off gradually with increasing substitution of Cr in the alloy. It nearly saturates to a value of 8 Å for $x \geq 0.4$. These values are indicated in Table I. Within the quantum version of Edwards and Anderson (EA) theory, Larsen has calculated T_f based on damping of the RKKY interaction due to a finite mean free path and fluctuations in the nearest-neighbor distance between the magnetic impurities.[22] In his theory, λ is determined from the expression:

$$\rho\lambda = \frac{[a_0^2 h(3/16\pi)^{1/3}]}{2\beta\, e^2}, \tag{2}$$

where a_0 is the lattice constant, ρ the total resistivity, and β an adjustable parameter which accounts for the difference between ordinary and transport cross section. The above expression has been used by some earlier workers, Sherlekar *et al.*,[14] Sheikh *et al.*,[21] and Cowen *et al.*[12] to calculate λ in *AuCu*Mn, *CuPt*Mn ternary spin-glass alloys. If one substitutes values of a_0 (=4.08 Å for pure Au), h and e, one obtains

$$\rho\lambda = 0.93 \times 10^{-11}/\beta\ \ \Omega\,\mathrm{cm}^2. \tag{3}$$

From the available data of T_f for *Au*Fe, Larsen has assumed a value of $\beta = 4$ which provided a satisfactory quantitative test of his theory. In the analysis of the data, he has shown that the theory fits satsifactorily even to *Au*Fe alloys showing RSG behavior (e.g., $Au_{83}Fe_{17}$). But if we substitute ρ values obtained in the present study to Eq. (3) assuming $\beta = 4$, then λ is found to vary from 5.56 Å for $Au_{82}Fe_{18}$ to 1.95 Å for $Au_{82}(Fe_{0.2}Cr_{0.6})_{18}$. These values of λ are very close to the lattice parameter and in some alloys even smaller than the near-neighbor distance ($\simeq$ 2.88 Å), which is physically impossible. A similar behavior has been found in *CuPt*Mn spin glasses by Cowen *et al.*[12] It therefore seems that the value of $\beta = 4$ assumed for the *Au*Fe system is not correct for high resistivity alloys as is the case in the present study. The physical interpretation has been questioned by several authors.[23–25] They have shown that the exponential decay over a distance (r) between the magnetic impurities greater than the electron mean free path (λ) results due to an averaging of the interaction over various impurity configurations and its application to the case of spin glasses, obscures the long-ranged nature of the RKKY interaction between the magnetic impurities. However, the observed T_f dependence is not wholly accounted for within the framework of these theories. In a recent work, Shegelski and Geldart[26,27] have tried to resolve this inadequacy by a consistent treatment of the *sd*-scattering and introducing new length scales in the effective spin-spin interaction. The authors have dealt with the case of two magnetic species[27] but they have omitted the quantitative effects and have provided only qualitative explanation. The validity of this theory in systems with high impurity concentration, specially near the percolation threshold like ours, remains to be examined.

IV. CONCLUSION

In the present study, we have reported the measurements of electrical resistivity in the ternary $Au_{82}(Fe_{1-x}Cr_x)_{18}$ system. The results show that the presence of Cr disrupts the ferromagnetic phase when $x > 0.1$ and leads to a cluster glass (CG) or spin-glass (SG) phase. The magnetic resistivity shows a maximum for $x \geq 0.6$ alloys which is probably due to the spin-glass phase of the alloy. The residual resistivity $\Delta\rho_0$ increases rapidly with increasing Cr concentration, saturating for Cr concentration $x \geq 0.4$. This could be due to the mean-free-path damping of the RKKY interaction. But there is no quantitative agreement with the theoretical predictions made by Larsen[22] because the application of this theory yields anomalously low values of the conduction electron mean free path λ. The alloys show a $T^{3/2}$ dependence of resistivity, the range of fit being larger for the cluster glass alloys. The reentrant systems ($0 \leq x \leq 0.1$) show a second region of $T^{3/2}$ dependence between 65 and 135 K indicating the presence of an additional scattering mechanism. At present, it is difficult to compare the data with the existing theories, since most of the theories in metallic systems assume that the spin-glass phase exists due to a dominant RKKY interaction between magnetic impurities. In the present alloys, magnetic impurity concentration is large enough to affect direct interaction between magnetic impurities and the various magnetic phases such as RSG, CG, and SG observed in the present alloys appear to be a consequence of competing ferro- and antiferromagnetic exchange interactions.

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