Resistivity minima in concentrated γ-$Cu_{100-x}Mn_x$ alloys ($36 \leqslant x \leqslant 83$)

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High-resolution electrical resistivity data of concentrated γ-$Cu_{100-x}Mn_x$ alloys with $x=36$, 60, 73, 76, and 83 have been presented here in the temperature range $1.2 \leqslant T \leqslant 30$ K. They show resistivity minima at T_{min} lying between 2.5 and 24.5 K. In this temperature range the alloys with $x=36$, 60, and 73 are cluster glasses while those with $x=76$ and 83 show a mixed cluster-glass and long-range antiferromagnetic phase. Resistivity below the minima follows a $\sqrt{T}$ type of behavior and has been interpreted in terms of the electron-electron (e-e) interaction effects in the presence of weak localization. The e-e interaction effects have dominant contributions to the resistivity in the temperature range of 2 K$\leqslant T \leqslant T_{min}/3$. The contributions from magnetic and phonon scattering are found to be negligible in this range. A good estimation of the density of states at the Fermi level, made from the coefficient of the $\sqrt{T}$ term, gives further support to the interpretation. In the higher temperature range of $T_{min}/3 \leqslant T \leqslant 30$ K, besides the e-e interaction effects, magnetic contribution of the type $T^{3/2}$ and phonon contribution given by the standard Bloch-Grüneissen relation have been observed. From our present findings and the earlier reports on other systems, we conclude that the $T^{3/2}$ type of magnetic contribution to the resistivity arises due to the low-temperature spin diffusive modes in spin/cluster glasses. The above analysis is insensitive to the magnetic state of the alloys.

I. INTRODUCTION

The resistivity minimum in metallic alloys has created a lot of interest in recent times. This was first observed at very low temperatures in dilute crystalline alloys[1] with magnetic impurity concentrations much less than 1 at. % and is known as Kondo effect. Later studies on metallic glasses[2,3] show resistivity minima at considerably higher temperatures compared to those for the dilute crystalline alloys. Some of the metallic glasses also show double minima.[4] A recent study by Das and Majumdar[3] has found a $\sqrt{T}$, T and $\sqrt{T}$ dependence of conductivity below minima at low, intermediate, and high temperatures in Co-rich amorphous alloys and this was interpreted in terms of weak localization and electron-electron (e-e) interaction effects. But for concentrated crystalline alloys there are only a few reports[5,6] on the resistivity minima. Interestingly, γ-phase (fcc) Cu-Mn alloys have always attracted very special attention due to their complex magnetic phases.[7,8] Resistivity studies have also shown some interesting features in different regions of Mn concentrations. Resistivity minima in dilute γ-CuMn alloys[1] has already been reported and was interpreted as Kondo effect whereas for concentrated alloys, only a rough estimate of the temperatures of the resistivity minima occurring around 20 K and depth of minima $[(\rho(1.2\text{ K})-\rho(T_{min}))/\rho(1.2\text{ K})]$ of less than 1% have been reported by Coles.[9] We have presented here very high resolution, dc-resistivity data for γ-phase concentrated $Cu_{100-x}Mn_x$ alloys ($x=36$, 60, 73, 76, and 83) in the temperature range $1.2 \leqslant T \leqslant 30$ K with the minima lying in the range of 2.5 to 24.5 K. The motivation behind the present study is to find out the physical phenomena responsible for the decrease in resistivity with increasing temperature below the minima in concentrated regime of this binary alloy system. In addition, magnetic and phonon scattering also have dominant contributions to the electrical resistivity in this temperature range. An attempt has been made here to estimate their individual contributions. This will help us in understanding the different competing phenomena resulting in the resistivity minima. Our measurements are restricted to 30 K only since the earlier work of Banerjee and Majumdar[7] had already covered, in the same alloy compositions as ours, the temperature range $30 \leqslant T \leqslant 300$ K.

II. EXPERIMENTAL DETAILS

The alloys were prepared by induction melting in pure argon atmosphere. Later they were heated to (900–950) °C for homogenization for at least 24 h and subsequently quenched fast to ice water to preserve their high-temperature γ phase (fcc) and also the random substitutional disorder. Homogeneity of those alloys are confirmed later by energy dispersive x-ray analysis (EDXA). A four-probe dc method was used to measure the electrical resistivity in a liquid He^4 cryostat with an automated data acquisition system through a personal computer using a GPIB (General Purpose Interface Bus) card. Data were taken at 25 mK interval or less in the temperature range below minima and 100 mK or higher in the temperature range above minima. The resolution of the present measurements ($\Delta\rho/\rho$ where ρ is the resistivity) is better than 5 ppm and the temperature stability is 3 to 50 mK depending on the range of temperature.

III. RESULTS AND DISCUSSION

Concentrated γ-phase $Cu_{100-x}Mn_x$ alloys with $x=36$, 60, 73, 76, and 83, studied by us, have exotic magnetic structures in the temperature range $1.2 \leqslant T \leqslant 30$ K. According to the magnetic phase diagram[7] they are cluster glasses for $x=36$, 60, and 73 with T_f between 135 and 149 K and are in the mixed cluster-glass and long-range antiferromagnetic phase for $x=76$ and 83 with $T_f \approx 145$ and 45 K, respectively. It will be rather interesting if one finds any dependence of

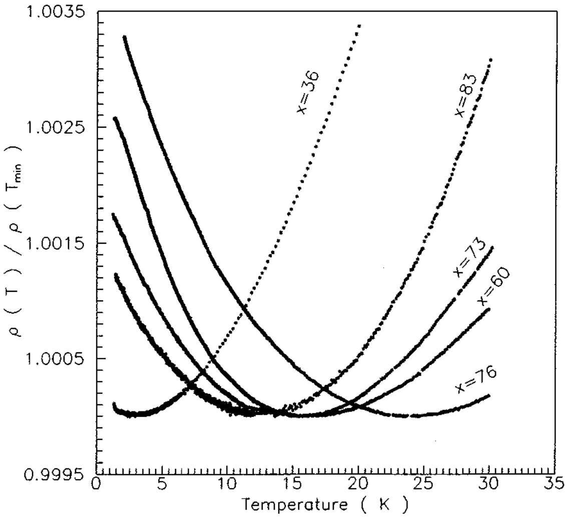

FIG. 1. Plot of the resistivity normalized with its value at T_{min} vs temperature for $\mathrm{Cu}_{100-x}\mathrm{Mn}_x$ alloys with $x=36$, 60, 73, 76, and 83 showing distinct minima.

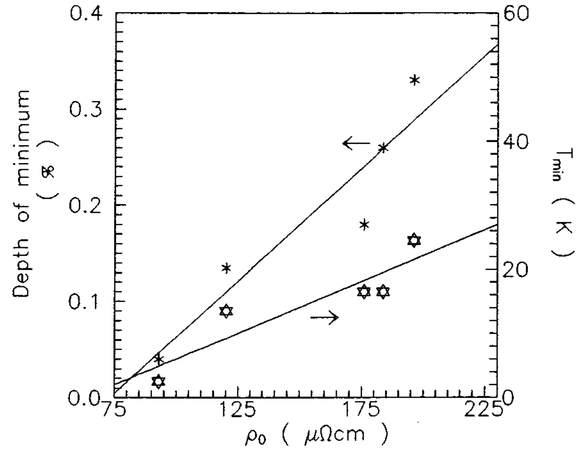

FIG. 2. Plot of the dependences of T_{min} and depth of minimum on the values of the residual resistivity of the alloys (see Table I).

the resistivity minima on their magnetic states. The present measurements on these alloys have shown minima in the range of 2.5 to 24.5 K with the depth of minima of the order of (0.04–0.33)% (details are given in Table I below). We have presented our raw data [$\rho(T)$ vs T] in Fig. 1. Here to get an expanded view of the minima, the plot for the alloy with $x=36$ has been shown till 20 K only whereas for the others they are shown till 30 K. It is interesting to note that the dispersion in the data is much less than the width of the symbols. The values of T_{min} reported by Coles[9] are in good agreement with those of the present investigation, but there it is claimed that no resistivity minima could be observed for $x<45$ at. %. However, we have a distinct minimum for $x=36$. The resistivity values at 1.2 K for all the alloys are in the range of (93–196) $\mu\Omega$ cm. These values differ by about 5% from those reported[7] for the same alloy compositions. A typical error of this order is generally found in the measurements of the thickness of the samples and the distance between the voltage probes. The large values of resistivity show that these are highly disordered materials where the resistivity increases with increasing Mn concentration until $x=76$ and then it drops in the Mn-rich $x=83$ which, according to Nordheim's rule, is quite expected. So no systematic dependence of T_{min} or the depth of the minima on alloy compositions has been found. However, correlations between the value of the resistivity with T_{min} and the depth of the minima have been observed. They show that the increasing value of resistivity shifts T_{min} to higher temperatures with higher depth of minima. On the other hand, more and more disorder introduced by varying the compositions of any alloy system will increase the value of resistivity. Hence it may be concluded that the increasing disorder in alloys can enhance the values of T_{min} as well as the depth of the minima. In Fig. 2, we have plotted them against the residual resistivity and found approximate linear relations in both the cases.

Now we shall examine the various physical phenomena which could describe the resistivity behavior below minima in $\mathrm{Cu}_{100-x}\mathrm{Mn}_x$ alloys with $x=60$, 73, 76, and 83. The alloy with $x=36$ has shown a minimum at 2.5 K. To find the functional dependence of the resistivity of this alloy in the temperature range below T_{min}, measurements have to be done much below 1.2 K which is not accessible to us. So we could not analyze the data of this alloy below T_{min}. However, from the analysis in the temperature range of $T \geqslant T_{\mathrm{min}}$, we have tried to find a plausible dependence of resistivity below T_{min}. In dilute crystalline alloys, according to the Kondo effect,[1] the decrease in resistivity with increasing temperature below minima follows the relation

$$\rho(T)=\rho_0-m\ \ln(T). \tag{1}$$

On the contrary, for highly disordered systems, the observed $\sqrt{T}$ dependence of resistivity below minima is generally interpreted in terms of the electron-electron interaction effects in the presence of weak localization.[10–12] This theory considers the phase coherence of two electrons both getting localized through elastic impurity scattering. The correction to the electrical conductivity,[10,11] $\Delta\sigma$, due to this e-e interaction effect goes as

$$\sigma(T)=\sigma_0+\Delta\sigma=\sigma_0+m_\sigma\sqrt{T}, \tag{2}$$

where

$$m_\sigma=\frac{1.3e^2}{4\sqrt{2}\pi^2\hbar}\left[\frac{4}{3}-\frac{3}{2}F_\sigma\right]\left[\frac{k_B}{\hbar D}\right]^{1/2}. \tag{3}$$

Here F_σ is the screening constant for Coulomb interactions and D is the diffusion constant. Earlier studies[2,3,5] on metallic glasses and concentrated crystalline alloys had shown a near-universal value of m_σ which is 6 $(\Omega\,\mathrm{cm}\,\mathrm{K}^{1/2})^{-1}$. The present alloys are very concentrated and thus it is very unlikely that they will behave as Kondo alloys. On the other hand, they are highly disordered and so the increase in resistivity below T_{min} may very well be attributed to the e-e interaction effects.[12] For convenience, in the present analysis, Eq. (2) has been modified from conductivity to resistivity as

$$\rho(T)=\rho_0+m_\rho\sqrt{T}, \tag{4}$$

where

TABLE I. Composition, values of depth of minimum $[(\rho(1.2\text{ K})-\rho(T_{min}))/\rho(1.2\text{ K})]$, T_{min}, parameters and χ^2 for fitting the data to Eq. (4) between 2 K and $T_{min}/3$ and to Eq. (9) between $T_{min}/3$ and 30 K.

($Cu_{100-x}Mn_x$) x (at. %)	Depth of minimum (%)	T_{min} (K)	Eq. (4) ρ_0 ($\mu\Omega$ cm)	Eq. (4) m_ρ $\left(\frac{\mu\Omega\text{ cm}}{\text{K}^{1/2}}\right)$	Eq. (4) χ^2 (10^{-10})	Eq. (9) ρ_0 ($\mu\Omega$ cm)	Eq. (9) m'_ρ $\left(\frac{\mu\Omega\text{ cm}}{\text{K}^{1/2}}\right)$	Eq. (9) B $\left(\frac{n\Omega\text{ cm}}{\text{K}^{3/2}}\right)$	Eq. (9) A ($\mu\Omega$ cm)	Eq. (9) χ^2 (10^{-10})
36	0.04	2.5				92.9	−0.05	5.5	77.1	4.7
60	0.18	16.5	176.0	−0.15	0.4	176.2	−0.23	4.9	27.5	1.1
73	0.26	16.5	183.7	−0.23	1.7	183.9	−0.40	8.0	29.2	1.7
76	0.33	24.5	196.4	−0.24	2.0	196.6	−0.35	4.2	81.0	0.3
83	0.14	13.5	120.1	−0.08	2.5	120.1	−0.13	3.2	482	8.2

$$m_\rho = -m_\sigma \rho_0^2, \quad (5)$$

assuming $m_\sigma \rho_0 \sqrt{T} \ll 1$ and so all the higher order terms of $\sqrt{T}$ are negligible in Eq. (4). Whether it is the Kondo effect or the interaction effects, they all occur at temperatures much below T_{min},[1,12] and hence the temperature range is chosen as $2\text{ K} \leqslant T \leqslant T_{min}/3$ in the present analysis. In this range our data have been fitted to both Eqs. (1) and (4). It is found that the value of the normalized χ^2 of the fit to Eq. (4) is an order of magnitude less than that to Eq. (1) for all the four samples. Here the normalized χ^2 has been defined as $(1/N)\Sigma_{i=1}^{N}[((\rho_{raw}^i-\rho_{fit}^i)^2)/\rho_{fit}^{i\,2}]$. The typical values of χ^2 are 1×10^{-9} and 1×10^{-10} for the $\ln(T)$ and $\sqrt{T}$ fits, respectively. The plot (not shown) of the deviation between the raw and the fitted data $(\rho_{raw}-\rho_{fit})$ with temperature for the $\ln(T)$ fit [Eq. (1)] describes the systematic trend whereas for the $\sqrt{T}$ fit [Eq. (4)] it is found to be random for all the alloys. This random nature of deviation can also be considered as a test for the goodness of the fit. Thus it is clear from the above discussion that the present data fit better to the $\sqrt{T}$ dependence of resistivity. The details of the fitting parameters with the values of χ^2 are given in Table I. The coefficient of the $\sqrt{T}$ term, i.e., m_ρ, in these alloys lies in the range (0.08–0.24) $\mu\Omega$ cm/K$^{1/2}$. The calculated values of m_σ [using Eq. (5)] are 4.8, 6.8, 6.2, and 5.6 $(\Omega\text{ cm K}^{1/2})^{-1}$ for $x=60$, 73, 76, and 83, respectively, and they are in very good agreement with the near-universal value of 6 $(\Omega\text{ cm K}^{1/2})^{-1}$.[2,3] A recent study on the electrical conductivity of Fe-rich FeNiCr system[6] below $T_{min}/2$ also found a $\sqrt{T}$ dependence but the values of m_σ are larger than the near-universal one. According to the generalized Einstein relation,[12] the resistivity is related to the density of states at the Fermi level, $N(E_F)$, and the diffusion constant, D, by

$$\rho = \frac{1}{e^2 N(E_F) D}. \quad (6)$$

On the other hand, m_σ is related to D by Eq. (3). So the value of $N(E_F)$ can be estimated from Eq. (6). Taking $F_\sigma=0$, the values of D, calculated from Eq. (3), are falling between (0.15–0.24) cm^2/sec. Hence the values of $N(E_F)$, obtained from Eq. (6) and using residual resistivity values from the fitting parameters, are in the range of $(1.4–2.6)\times10^{35}$ erg^{-1}cm^{-3}. An earlier specific heat study[13] on CuMn had shown that the alloys under the present investigation have their electronic specific heat coefficient (γ) of the order of 10 mJ/mol K^2. Thus the value of $N(E_F)$, calculated from γ using the free-electron theory relation, $N(E_F)=3\gamma/\pi^2 K_B^2$ is 2.2×10^{35} erg^{-1}cm^{-3}. This shows that the values of the density of states, obtained in the present work, agree well with those calculated from the experimentally obtained electronic specific heat coefficient.[13] So a good estimation of the density of states at the Fermi level can certainly be made from m_ρ. Hence a $\sqrt{T}$ dependence of the resistivity in the temperature range below minima, interpreted as coming from e-e interaction effects, is well justified here in these concentrated $Cu_{100-x}Mn_x$ alloys. On the other hand, the Kondo effect gives a better description of the resistivity behavior below minima in the dilute regime of this binary alloy system.[1] Therefore CuMn is a unique alloy system where the resistivity minima can be described by both the Kondo and the e-e interaction effects depending on the concentration regime.

The analysis of $\rho(T)$ in the temperature range $T_{min}/3 \leqslant T \leqslant 30$ K is presented below. Since $x=36$ is a rather concentrated alloy with a strong disorder $[\rho(1.2\text{ K})=92.8$ $\mu\Omega$ cm], one can expect the e-e interaction effects to be responsible for its resistivity minimum as it has already been observed in the case of alloys with higher Mn concentrations in the present investigation. Besides the e-e interaction effects, one also expects the contributions to the measured resistivity from other competing effects. Phonon contribution, however small it might be at low temperatures, is always present. In addition, the effect of cluster-glass type of magnetic order of the present alloy system will have sufficient magnetic contribution to the resistivity. So the measured resistivity, assuming Matthiessen's rule, is the sum of all those contributions given by

$$\rho(T)=\rho_0+\rho_{interaction}(T)+\rho_{phonon}(T)+\rho_{magnetic}(T), \quad (7)$$

where ρ_0 is the residual resistivity. For phonon contribution, we have taken the standard Bloch-Grüneissen relation

$$\rho_{phonon}(T)=A\left(\frac{T}{\theta_D}\right)^5 \int_0^{\theta_D/T} \frac{z^5\,dz}{(e^z-1)(1-e^{-z})}, \quad (8)$$

where A is a constant and θ_D is the Debye temperature. At very low temperatures (much below the spin-freezing temperature, T_f), magnetic contribution to the resistivity arising from the scattering of conduction electrons by the spin-diffusive modes in spin/cluster glasses is proportional to $T^{3/2}$, as proposed by Rivier and Adkins.[14] Later Fischer[15] suggested a $(BT^2-CT^{5/2})(B,C>0)$ type of dependence of resistivity at low temperatures $(T<T_f)$. In this model, the scattering of conduction electrons by the low-energy spin excitations along with the static disorder of impurity spins was considered. So the final expressions for the resistivity become

$$\rho(T)=\rho_0+m'_\rho\sqrt{T}+BT^{3/2} + A\left(\frac{T}{\theta_D}\right)^5 \int_0^{\theta_D/T} \frac{z^5\,dz}{(e^z-1)(1-e^{-z})} \tag{9}$$

and

$$\rho(T)=\rho_0+m'_\rho\sqrt{T}+BT^2-CT^{5/2} + A\left(\frac{T}{\theta_D}\right)^5 \int_0^{\theta_D/T} \frac{z^5\,dz}{(e^z-1)(1-e^{-z})}. \tag{10}$$

The values of θ_D for $x=36$, 60, 73, 76, and 83, taken from an earlier report,[7] are 325, 305, 305, 325, and 360 K, respectively. First we have fitted the data to Eq. (9) and found that they fit very well and the normalized value of χ^2 of the order of 1×10^{-10} is consistent with our experimental accuracy. All the details of the fit are given in Table I. On the other hand, fitting to Eq. (10) gives unphysical signs to some of the parameters for all the alloys. The above findings show conclusively that the $T^{3/2}$ type of magnetic contribution along with ρ_0, lattice and e-e interaction effects give the best description of the resistivity in the temperature range between $T_{\min}/3$ and 30 K. But the high-temperature $(T>30$ K) resistivity study by Banerjee and Majumdar[7] found the $(BT^2-CT^{5/2})(B,C>0)$ type of magnetic contribution in the same alloy compositions. They had interpreted the data in terms of the diffusive spin excitations as the dominant source of electron scattering. According to Fischer,[15] $(BT^2-CT^{5/2})$ type of magnetic contribution is valid in the temperature range where the Kondo effect is negligible. This certainly indicates that it is applicable at sufficiently high temperatures above the resistivity minima. It was also shown that instead of $(BT^2-CT^{5/2})$, a $T^{3/2}$ type of magnetic contribution arises due to the ferromagnetic clusters in spin glasses at temperatures well above minima. However, in CuMn binary alloys the clusters are predominantly antiferromagnetic. On the contrary, the magnetic contribution of $T^{3/2}$ type, as suggested by Rivier and Adkins,[14] has its effects in the resistivity at low temperatures $(T\ll T_f)$,[16] where resistivity minima are generally found. Therefore both $T^{3/2}$ (Rivier and Adkins, $T\ll T_f$) and $(BT^2-CT^{5/2})$ or $T^{3/2}$ (both Fischer, $T<T_f$) type of contributions to the resistivity may be expected at different temperature regions in the same alloy compositions where minima occur at temperatures much below T_f. Hence the earlier findings[7] of $(BT^2-CT^{5/2})$ type of dependence above 30 K in concentrated CuMn alloys where $T_{\min}\sim20$ K are quite justified. Another study by Ford and Mydosh[16] had found a $T^{3/2}$ type of magnetic contributions in $Cu_{100-x}Mn_x$ alloys with $x\leqslant11$ at. % and also in AuCr, AuMn, and AgMn systems. There the temperature range of the $T^{3/2}$ fit was 1.5 K$\leqslant T\leqslant T_f/4$. The temperature range of the present measurements, $1.2\leqslant T\leqslant30$ K, is below $T_f/4$ (except for the alloy, $x=83$, with $T_f=45$ K) and this agrees with the range of study of Mydosh and Ford. One interesting point, to be noted from the present findings in concentrated CuMn alloys and also from the earlier report by Ford and Mydosh,[16] is that the magnetic contribution of the type $T^{3/2}$ (Rivier and Adkins) is observed in spin glasses only at low temperatures, generally below $T_f/4$. The coefficient B of the $T^{3/2}$ term, according to Rivier and Adkins,[14] should have dependence on the magnetic impurity concentration. Earlier resistivity study[16] on $Cu_{100-x}Mn_x$ with $x\leqslant11$ at. % had shown the dependence of B on x. In our case no systematic dependence of B on x has been found (Fig. 3). But the values of B obtained here are in the vicinity of (3.2–8) $(n\Omega)$cm K$^{-3/2}$ which agrees with 7.7 $(n\Omega)$cm K$^{-3/2}$ for the $Cu_{90.3}Mn_{9.7}$ alloy.[16]

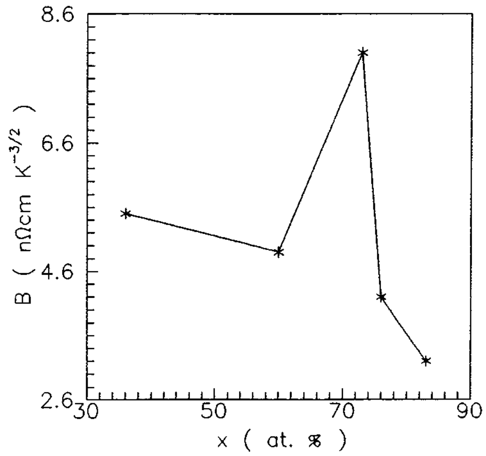

FIG. 3. Plot of B (coefficient of the magnetic contributions) of Eq. (9) vs Mn concentration, x, in $Cu_{100-x}Mn_x$ alloys.

In Fig. 4, we have plotted the individual contributions to the resistivity from magnetic, phonon, and e-e interactions along with the fitted (sum of all the contributions) and the raw $[\Delta\rho=\rho(T)-\rho_0]$ data. Here the fit is so good that the raw and the fitted data are indistinguishable. Moreover, the fits

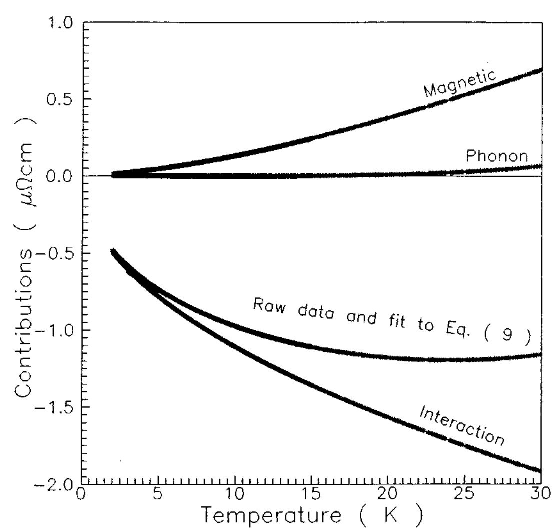

FIG. 4. Plot of magnetic $(T^{3/2})$, phonon, and electron-electron interaction $(\sqrt{T})$ contributions along with the raw data $[\rho(T)-\rho_0]$ and the fit to Eq. (9) vs temperature for the alloy with $x=76$.

seem to be independent of the detailed magnetic state of the alloys, although the cluster-glass phase is common to all of them. Below $T_{min}/3$ the magnetic and phonon contributions are so small that it is enough to consider the contribution from the interaction effects only, besides ρ_0. The typical values at 8 K for phonon, magnetic, and interaction contributions are 2×10^{-5}, 1×10^{-1}, and 1 (all are in $\mu\Omega$ cm), respectively, for the alloy with $x=76$. At still lower temperatures the values of phonon and magnetic contributions are much smaller compared to that due to the interaction effects. This can be seen in Fig. 4. Therefore the choice of $T_{min}/3$ as the upper limit in the low temperature analysis is quite justified. It is to be noted here that the $\sqrt{T}$ contribution due to the interaction effect should ideally have the same coefficient for both ranges of temperature (2 K$\leqslant T\leqslant T_{min}/3$ and $T_{min}/3\leqslant T\leqslant 30$ K). That is why we have chosen to fit the resistivity rather than the conductivity in the 2 K$\leqslant T\leqslant T_{min}/3$ range. The values of m'_ρ are in excellent agreement with the values of m_ρ considering the fact that the former is obtained along with the residual resistivity, phonon, and magnetic contributions in the temperature range $T_{min}/3\leqslant T\leqslant 30$ K, whereas m_ρ is obtained along with only the residual resistivity in the range of 2 K$\leqslant T\leqslant T_{min}/3$. The values differ in the two cases by about only 60% and this is quite reasonable with so much of variations in the range of temperatures and the fitting parameters. For $x=36$, the calculated value of m_σ from the value of m'_ρ using Eq. (5) (replacing m_ρ by m'_ρ) is 6.37 $(\Omega\text{ cm K}^{1/2})^{-1}$ and it is found to be almost equal to the near-universal value of 6 $(\Omega\text{ cm K}^{1/2})^{-1}$.

IV. CONCLUSIONS

In conclusion, a distinct $\sqrt{T}$ dependence of resistivity below minima has been found in the concentrated $Cu_{100-x}Mn_x$ alloys with $x=36$, 60, 73, 76, and 83 in the range 2 K$\leqslant T\leqslant T_{min}/3$. Linear correlations are obtained between the depth of the minima as well as the T_{min} and the resistivity of the alloys. A good estimation of the density of states at the Fermi level has been made from the coefficient of the $\sqrt{T}$ term. The value is in good agreement with that obtained from specific heat measurements. Above $T_{min}/3$ and to 30 K, the magnetic contribution of the $T^{3/2}$ type has been found along with those from phonon and interaction effects. It is also concluded that the $T^{3/2}$ contribution to the resistivity due to the spin diffusive modes in spin/cluster glasses is observed at low temperature ($T\leqslant T_f/4$) whereas ($BT^2-CT^{5/2}$) type of magnetic contribution is found at much higher temperatures compared to T_{min}. This shows the simultaneous presence of the $T^{3/2}$ type of magnetic contribution along with that from the e-e interaction effects. It also reveals that both $T^{3/2}$ and $(BT^2-CT^{5/2})(B,C>0)$ terms can be observed in the same alloys in different temperature ranges. The above conclusions are found to be independent of the details of the magnetic state of the alloys.

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