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## Resistivity and Magnetoresistance of Dilute Solutions of Mn in Cu-Ni Alloys\*

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(Received 4 August 1971)

The electrical resistivity from 2 to 100 K and the longitudinal magnetoresistance from 0 to 85 kOe at 4.2 K have been measured on  $\text{Cu}_{1-x}\text{Ni}_x$  alloys ( $x=0.06, 0.12, 0.23$ ) containing 0 to 1175 ppm Mn as magnetic impurities. All Mn-bearing samples exhibited resistivity minima, and the difference in resistivity between each  $\text{Cu}_{1-x}\text{Ni}_x(\text{Mn})$  alloy and its Mn-free equivalent depended linearly on  $\log_{10}T$  for nearly an order of magnitude in  $T$ . These phenomena are characteristic of dilute magnetic alloys and suggest that the results can be interpreted in terms of the Kondo effect. This picture is supported by the magnetoresistance of the alloys, which was negative for all Mn-bearing samples. The magnetic contributions to the resistivity and magnetoresistance were also proportional to the Mn concentration and essentially independent of Ni concentration. The former result is interpreted as an indication that interaction effects among the Mn ions were negligible, and the latter result suggests that the Ni had very little effect on the local-moment character of the Mn.

### I. INTRODUCTION

The resistivity minimum of dilute magnetic alloys was explained theoretically by Kondo.<sup>1</sup> He based his theory on a model in which it was assumed that localized magnetic moments form at the impurity sites and subsequently interact with the conduction electrons via the  $s$ - $d$  exchange interaction. His finding stimulated a considerable

amount of theoretical and experimental interest in the properties of dilute magnetic alloys. (Excellent reviews of the theoretical and experimental situations up to 1969 have been given by Kondo<sup>2</sup> and Heeger,<sup>3</sup> respectively.) One of the metals which has been used quite extensively as the host in the experimental work on this problem is Cu. A wealth of evidence<sup>3,4</sup> indicates that Fe, Mn, and Cr exhibit local-moment character in Cu, and of

particular interest, with respect to the work discussed in this paper, are the experiments of Monod<sup>5</sup> and Daybell and Steyert<sup>6,7</sup> which show that dilute amounts of Mn, Fe, and Cr in Cu give rise to resistivity minima. In addition, negative magnetoresistance<sup>5,7,8</sup> is observed in these alloys, and this is in qualitative agreement with theoretical predictions based on the *s-d* model.<sup>9-11</sup>

The above-mentioned experiments with Cu as the host form a good background from which to study how changing the host in some systematic fashion alters the properties of the alloy which are attributable to the interaction between the host and the impurities. This paper reports about one such study<sup>12</sup> in which the Cu host was changed by alloying with up to 23-at. % Ni and using Mn in concentrations up to 1175 at. ppm as the magnetic impurity. Although Cu and Ni ideally form solid solutions throughout the entire composition range, only  $\text{Cu}_{1-x}\text{Ni}_x$  alloys with  $x \leq 0.23$  ( $x$  denotes the atomic fraction of Ni in a given alloy) were used, because for Ni concentrations much greater than 23% anomalous behavior has been observed. In particular, resistivity minima<sup>13-15</sup> and maxima,<sup>16</sup> as well as negative magnetoresistance,<sup>13,14</sup> have been observed for alloys with  $0.3 \leq x \leq 0.47$ , and these phenomena have been attributed to magnetic polarization clouds which are associated with small Ni clusters in the host.<sup>14,16</sup> In addition, for  $x \geq 0.47$ ,  $\text{Cu}_{1-x}\text{Ni}_x$  alloys are ferromagnetic. On the other hand, Ryan *et al.*<sup>17</sup> and Seib and Spicer<sup>18</sup> indicate that for  $x \leq 0.23$ , the alloys should be well behaved and the Ni clustering should be of little consequence, provided that the alloys are given the proper heat treatment. In addition to these considerations, the Cu-Ni system was also chosen so that a comparison could be made between the present work and a similar investigation carried out by Gärtner *et al.*<sup>13,19,20</sup> on a series of Cu-Ni(Fe) alloys.

## II. EXPERIMENTAL PROCEDURE

The alloys used in this study were prepared from 99.99%-pure Cu and 99.999%-pure Ni which were supplied by the American Smelting and Refining Company and Atomergic Chemetals Company, respectively. The Cu and Ni were melted in an electron-beam furnace under high vacuum to remove volatile impurities and dissolved gases in the as-received metals. The Mn was supplied by Schmidt of this laboratory who purified some commercial grade electrolytic Mn by means of a sublimation technique. From these starting materials, three  $\text{Cu}_{1-x}\text{Ni}_x$  master alloys with  $x = 0.06$ , 0.12, and 0.23 were prepared by arc-melting together the appropriate quantities of the constituents in a shallow graphite cup which allowed the copper to become hotter and completely dissolve the nickel.

A master alloy containing 1-at. % Mn was prepared by arc-melting, and each alloy was formed by arc-melting together on a cold copper hearth a piece of a given  $\text{Cu}_{1-x}\text{Ni}_x$  master alloy and the required amount of the Mn master alloy to give the desired Mn concentration. Then, this arc-melted button was swaged and drawn through a tungsten carbide die. The resulting 0.040-in.-diam wire was electropolished in a nitric-acid-methanol electrolyte to remove any surface contamination and provide a bright clean surface. 1-in. lengths of the wire were sealed in evacuated quartz ampoules, given a homogenizing anneal at 1000 °C for 3 days, and were quenched in ice water.

The resistivity measurements were made by the standard dc four-probe method using a Honeywell six-dial potentiometer and a Guildline photocell galvanometer amplifier. This system has a resolution of  $\pm 0.01 \mu\text{V}$ , and since typical readings were on the order of 200  $\mu\text{V}$ , the voltage drops across the samples were measured to five significant figures. A constant current supply which was built in this laboratory provided the current, and it was stable to one part in  $10^5$  during the time required for a given measurement. The temperature of the samples was measured with Au-Fe-vs-Cu and constantan-vs-Cu thermocouples in the ranges 2-25 K and 25-100 K, respectively. The longitudinal magnetoresistance of the samples was measured at 4.2 K in magnetic fields up to 85 kOe using the system described above and an RCA model SM2804 superconductive magnet.

## III. RESULTS

The electrical resistivity  $\rho$  of the alloys listed in Table I was measured as a function of temperature from 2 to 100 K. The data on the 6-at. % Ni samples are representative of these measurements and are shown in Fig. 1. Each one of the Mn-bearing samples exhibits a resistivity minimum, and below the temperature  $T_{\text{min}}$  at which the minimum occurs, the resistivity continues to increase

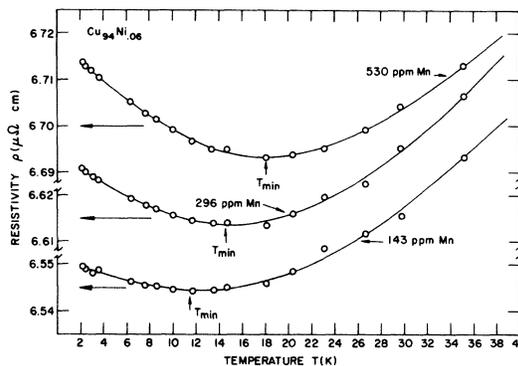


FIG. 1. Resistivity of the  $\text{Cu}_{0.94}\text{Ni}_{0.06}$  (Mn) alloys.

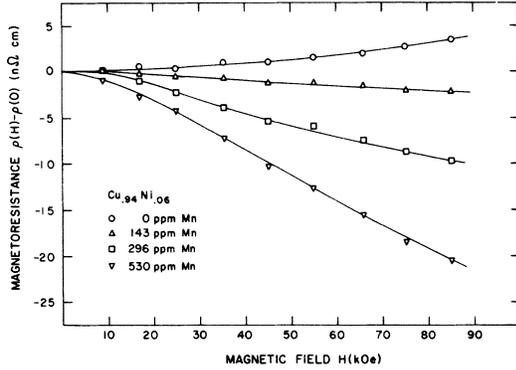


FIG. 2. Magnetoresistance of the  $\text{Cu}_{0.94}\text{Ni}_{0.06}$  (Mn) alloys.

to the lowest temperatures reached in the experiment. This anomalous behavior can be characterized by the depth of the minimum which is taken to be  $\rho(2.2 \text{ K}) - \rho(T_{\text{min}})$  for definiteness. Values of  $T_{\text{min}}$  and  $\rho(2.2 \text{ K}) - \rho(T_{\text{min}})$  are given in Table I for all of the Mn-bearing samples.

The resistivity was also measured at 4.2 K as a function of longitudinal magnetic field from 0 to 85 kOe. Figure 2 shows the magnetoresistance  $\rho(H) - \rho(0)$  of the 6% Ni samples. The results on the 12 and 23% Ni samples are similar and are not shown, but values of the magnetoresistance of all the samples at 85 kOe have been listed in Table I for comparison. The magnetoresistance of the Mn-free samples behaves in the "normal" fashion in that it increases with increasing field, but the magnetoresistance of the Mn-bearing alloys is negative. This anomalous behavior can be characterized by the quantity  $\Delta\rho(H)$ , which is defined to be the difference between the magnetoresistance of a given Mn-bearing alloy and its corresponding Mn-free equivalent; values of  $\Delta\rho(85 \text{ kOe})$  are

listed in Table I for all of the samples.

The data presented in Figs. 1 and 2 indicate that the anomalous behavior of the Cu-Ni(Mn) alloys scales with Mn concentration. This idea is supported by the fact that the values of  $\rho(2.2 \text{ K}) - \rho(T_{\text{min}})$  and  $\Delta\rho(85 \text{ kOe})$  listed in Table I are roughly proportional to the Mn concentration; to show this more explicitly,  $[\rho(2.2 \text{ K}) - \rho(T_{\text{min}})]/\text{ppm Mn}$  and  $\Delta\rho(85 \text{ kOe})/\text{ppm Mn}$  are also given in Table I. Considering the uncertainty<sup>21</sup> of 10–15% in the Mn concentrations listed in Table I, the values of  $[\rho(2.2 \text{ K}) - \rho(T_{\text{min}})]/\text{ppm Mn}$  and  $\Delta\rho(85 \text{ kOe})/\text{ppm Mn}$  are essentially independent of the Mn concentration; this suggests that the behavior of the alloys discussed in this paper can be interpreted in terms of the sum of effects due to noninteracting impurities.

#### IV. DISCUSSION

##### A. Resistivity

The early theory of dilute magnetic alloys given by Kondo<sup>1,2</sup> predicts that the impurity contribution to the resistivity  $\Delta\rho(T)$  should be given by

$$\Delta\rho(T) = BcJ^2S(S+1)[1 + 4J\rho_1 \ln(k_B T/D)] \quad (1)$$

where  $B$  is a constant for a given host,  $c$  is the atomic concentration of magnetic impurities,  $J$  is the strength of the  $s$ - $d$  interaction,  $S$  is the spin on the impurity,  $\rho_1$  is the density of states per atom at the Fermi level for one direction of the electron spin,  $k_B$  is Boltzmann's constant, and  $D$  is the half-width of the conduction band. Thus, if Kondo's theory is applicable to our results, a plot of  $\Delta\rho(T)/c$  vs  $\ln(k_B T/D)$  should be a straight line with a slope  $m$  given by

$$m = 4BJ^3S(S+1)\rho_1 \quad (2)$$

Assuming Matthiessen's rule is valid for the alloys

TABLE I. Values of the temperature at which the minimum occurs,  $T_{\text{min}}$ ; the depth of the minimum,  $\rho(2.2 \text{ K}) - \rho(T_{\text{min}})$ ; the magnetoresistance at 85 kOe,  $\rho(85 \text{ kOe}) - \rho(0)$ ; the impurity contribution to the magnetoresistance at 85 kOe,  $\Delta\rho(85 \text{ kOe})$ ; the depth of the minimum per ppm Mn; and  $\Delta\rho(85 \text{ kOe})/\text{ppm Mn}$ .

Sample (at. ppm Mn)	$T_{\text{min}}$ (K)	$\rho(2.2 \text{ K}) - \rho(T_{\text{min}})$ ( $n\Omega \text{ cm}$ )	$\rho(85 \text{ kOe}) - \rho(0)$ ( $n\Omega \text{ cm}$ )	$\Delta\rho(85 \text{ kOe})$ ( $n\Omega \text{ cm}$ )	$\frac{\rho(2.2 \text{ K}) - \rho(T_{\text{min}})}{\text{ppm}}$	$\frac{\Delta\rho(85 \text{ kOe})}{\text{ppm}}$
$\text{Cu}_{0.94}\text{Ni}_{0.06}$ (0 ppm Mn)	...	...	3.6	...	...	...
$\text{Cu}_{0.94}\text{Ni}_{0.06}$ (143 ppm Mn)	11.5	5.3	-1.9	-5.5	0.037	-0.039
$\text{Cu}_{0.94}\text{Ni}_{0.06}$ (296 ppm Mn)	14.6	12.5	-9.6	-13.2	0.042	-0.045
$\text{Cu}_{0.94}\text{Ni}_{0.06}$ (530 ppm Mn)	18.1	20.3	-20.4	-24.0	0.038	-0.045
$\text{Cu}_{0.88}\text{Ni}_{0.12}$ (0 ppm Mn)	...	...	2.6	...	...	...
$\text{Cu}_{0.88}\text{Ni}_{0.12}$ (156 ppm Mn)	14.8	5.1	-4.7	-7.3	0.033	-0.047
$\text{Cu}_{0.88}\text{Ni}_{0.12}$ (269 ppm Mn)	16.8	12.3	-10.9	-13.5	0.046	-0.051
$\text{Cu}_{0.88}\text{Ni}_{0.12}$ (570 ppm Mn)	21.4	25.7	-23.2	-25.8	0.045	-0.045
$\text{Cu}_{0.77}\text{Ni}_{0.23}$ (0 ppm Mn)	...	...	2.0	...	...	...
$\text{Cu}_{0.77}\text{Ni}_{0.23}$ (263 ppm Mn)	18.9	13.0	-11.6	-13.6	0.049	-0.051
$\text{Cu}_{0.77}\text{Ni}_{0.23}$ (674 ppm Mn)	20.9	23.2	-23.4	-25.4	0.034	-0.038
$\text{Cu}_{0.77}\text{Ni}_{0.23}$ (1175 ppm Mn)	23.5	39.6	-44.5	-46.5	0.034	-0.039

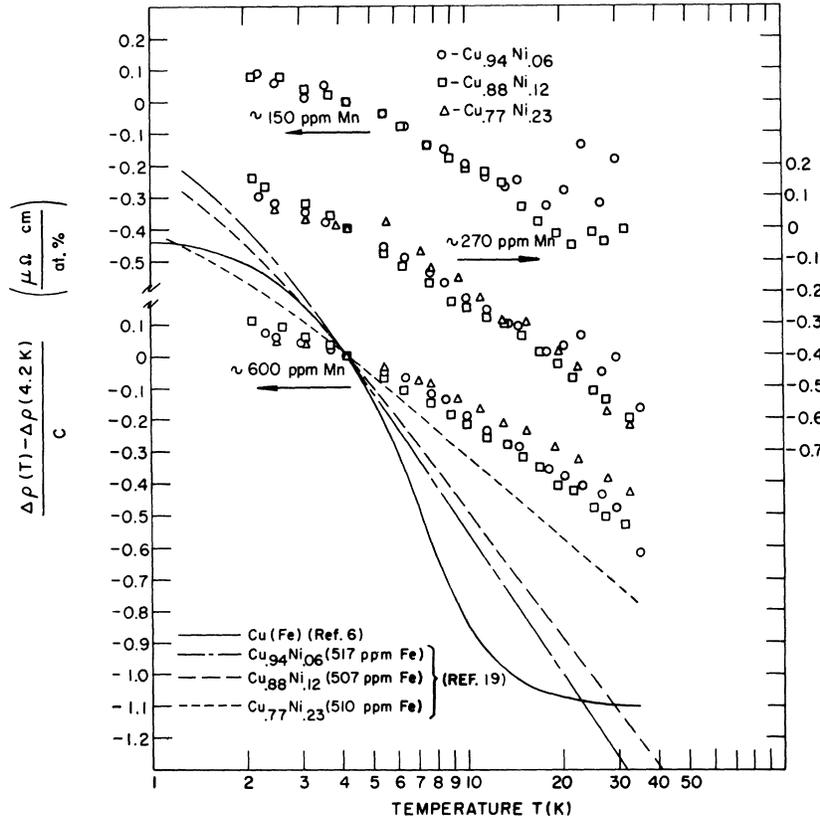


FIG. 3. Impurity contribution to the resistivity  $\Delta\rho(T)$  per at. % Mn normalized to the 4.2 K value vs  $\log_{10} T$  for the  $\text{Cu}_{1-x}\text{Ni}_x(\text{Mn})$  and the  $\text{Cu}_{1-x}\text{Ni}_x(\text{Fe})$  alloys.

in question,  $\Delta\rho(T)$  is given by the difference between the resistivity of a Mn-bearing sample and its Mn-free equivalent. Figure 3 shows the results of this subtraction where values of  $[\Delta\rho(T) - \Delta\rho(4.2\text{ K})]/c$  are plotted vs  $\log_{10} T$ . The data are normalized to 4.2 K in order to account for errors in the geometry of the samples and are presented such that alloys with roughly the same Mn concentration but varying amounts of Ni are plotted together. Between about 4 and 20 K,  $[\Delta\rho(T) - \Delta\rho(4.2\text{ K})]/c$  depends linearly on  $\log_{10} T$ , in agreement with Kondo's theory, and values of  $m$  determined from the straight-line portions of the curves in Fig. 3 are listed in Table II. Below about 4 K the curves begin to bend over, and Kondo's theory is apparently no longer applicable. However, more sophisticated theories<sup>22-24</sup> based on the  $s$ - $d$  interaction indicate that Kondo's perturbation calculation breaks down below the Kondo temperature  $T_K$  which is given by

$$k_B T_K \sim D e^{-1/|J|\rho_1}, \quad (3)$$

and a many-body singlet ground state is formed in which the conduction electrons compensate the impurity spins. In the alloys under discussion here, extra conduction-electron scattering is introduced by the Ni and this very likely affects the magnetic contribution to the low-temperature

resistivity, as suggested by Gainon and Heeger<sup>25</sup> and by Suhl.<sup>26</sup> Hence, it is difficult to determine concise Kondo temperatures but, qualitatively, it appears that our results, as seen in Fig. 3, have a  $T_K$  which is not widely different from the 0.2 K value found for Cu(Mn).<sup>4,5</sup>

The most striking feature of the data is that the behavior of the Mn-bearing samples is so similar. Taking the uncertainty in the Mn concentration into account, it can be concluded from Fig. 3 and Table II that the data are essentially independent of both the Mn concentration and the Ni concen-

TABLE II. Values of the slopes  $m$  and  $m'$  for the Mn-bearing alloys.

Sample (at. ppm Mn)	$m$ ( $\mu\Omega\text{ cm/at.}\%$ )	$m'$ ( $10^{-3}\pi\Omega\text{ cm}/$ kOe ppm)
Cu <sub>0.94</sub> Ni <sub>0.06</sub> (143 ppm Mn)	-0.26	-0.55
Cu <sub>0.94</sub> Ni <sub>0.06</sub> (296 ppm Mn)	-0.28	-0.58
Cu <sub>0.94</sub> Ni <sub>0.06</sub> (530 ppm Mn)	-0.27	-0.61
Cu <sub>0.88</sub> Ni <sub>0.12</sub> (156 ppm Mn)	-0.29	-0.58
Cu <sub>0.88</sub> Ni <sub>0.12</sub> (269 ppm Mn)	-0.28	-0.67
Cu <sub>0.88</sub> Ni <sub>0.12</sub> (570 ppm Mn)	-0.25	-0.58
Cu <sub>0.77</sub> Ni <sub>0.23</sub> (263 ppm Mn)	-0.31	-0.70
Cu <sub>0.77</sub> Ni <sub>0.23</sub> (674 ppm Mn)	-0.21	-0.49
Cu <sub>0.77</sub> Ni <sub>0.23</sub> (1175 ppm Mn)	-0.20	-0.51

tration. The former result gives added support to the contention that the alloys are, in fact, dilute, and the latter suggests that the Ni has very little effect on the local-moment character of the Mn.

This latter result is surprising in view of the fact that Gärtner *et al.*<sup>19</sup> found a rather strong dependence of  $\Delta\rho(T)$  on Ni concentration in a series of  $\text{Cu}_{1-x}\text{Ni}_x(\text{Fe})$  alloys. Their data on samples with a nominal Fe concentration of 500 at. ppm, the data of Daybell and Steyert<sup>6</sup> on the Cu(Fe) system, and our results are shown in Fig. 3 for comparison; the decreasing slopes of the lines with increasing Ni content are evident in the Fe-bearing samples. In fact, the value of  $m$  decreases by roughly a factor of 2 as the Ni concentration increases from 0 to 23% in the Fe-bearing alloys, whereas the slope varies only slightly in the case of the Mn-bearing samples. One hypothesis that Gärtner *et al.*<sup>19</sup> put forth to explain their results was that Fe was removed from participation in the Kondo effect because the Fe either nucleated or otherwise became associated with small Ni clusters in the host. This view was supported by the Mössbauer work of Bennett *et al.*<sup>27</sup> who showed that a small Ni cluster was associated with an isolated Fe atom in  $\text{Cu}_{1-x}\text{Ni}_x$  alloys. Our work indicates that if this type of phenomenon is occurring in the Mn-bearing samples, it is not nearly as pronounced as in the case of the Fe-bearing alloys.

#### B. Magnetoresistance

The effects of an external magnetic field on the properties of dilute magnetic alloys have been investigated by several workers<sup>9-11</sup> and the picture which has evolved from these theories is that the scattering of the conduction electrons by the impurity spins should be suppressed in a magnetic field. Consequently, at a given temperature the resistivity should decrease as the magnetic field increases and eventually saturate or become only weakly field dependent in large magnetic fields. This behavior is related to the "freezing out" of the impurity spins by the magnetic field and a subsequent reduction in the spin-flip scattering processes responsible for the Kondo effect. Once the spins are frozen out, Rohrer<sup>28</sup> suggests that higher-order scattering processes involving no net spin flip of the impurity govern the magnetoresistance, and this leads to only weakly field-dependent behavior. Assuming that the populations of the Zeeman levels of the impurities are governed by the Boltzmann factor, Béal-Monod and Weiner<sup>9</sup> indicate that saturation should occur for fields which satisfy the relation

$$g \mu_B H / k_B T > 4, \quad (4)$$

where  $g$  is the gyromagnetic ratio of the impurity spins (usually taken to be 2) and  $\mu_B$  is the Bohr

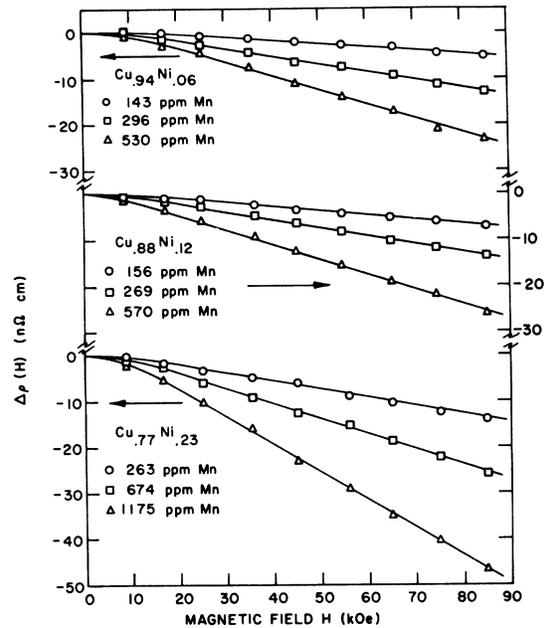


FIG. 4. Impurity contribution to the magnetoresistance  $\Delta\rho(H)$  vs  $H$  for the  $\text{Cu}_{1-x}\text{Ni}_x(\text{Mn})$  alloys.

magneton. For  $T = 4.2$  K, Eq. (4) implies that the magnetoresistance should saturate for  $H > 60$  kOe.

In order to compare our results with the theoretical predictions, some technique must be adopted for separating the "normal" positive magnetoresistance of the dilute magnetic alloys from the negative contribution we have been discussing. As far as the authors have been able to determine, there is no general method for performing this separation. Various techniques have been discussed in the literature,<sup>5,28,29</sup> and the differences between them reflect the inherent difficulty in interpreting the magnetoresistance of dilute magnetic alloys. The method that the authors have chosen is to assume that the magnetoresistance of a  $\text{Cu}_{1-x}\text{Ni}_x$  master alloy is a measure of the normal positive magnetoresistance of its corresponding Mn-bearing alloys, i. e., that the magnetoresistance of the master alloys is about what the magnetoresistance of the Mn-bearing alloys would be if the Mn were nonmagnetic. Since the Ni itself so strongly scatters the electrons, it seems reasonable to assume that a few hundred ppm of a nonmagnetic impurity would have little effect on the magnetoresistance of the Cu-Ni hosts. Figure 4 shows the results of this separation where the quantity  $\Delta\rho(H)$ , which is defined to be the difference between the magnetoresistance of a Mn-bearing alloy and its Mn-free equivalent, is plotted vs magnetic field. Comparing these results with the theoretical predictions, it can be seen that the ex-

pected saturation in high fields does not occur. In fact, above about 25 kOe,  $\Delta\rho(H)$  varies linearly with field. However, the results are again independent of both the Mn concentration and the Ni concentration. The slopes per ppm Mn,  $m'$ , of the straight-line portions of the curves in Fig. 4 are listed in Table II, and considering the uncertainty in the Mn concentration, they are roughly the same for all of the samples.

Because of the nonsaturation behavior of the magnetoresistance, direct comparison with theoretical expressions is difficult, but the results can be compared with experiments on related systems. Rohrer<sup>8</sup> investigated the magnetoresistance of dilute Cu(Mn) and Cu(Fe) alloys, and he found that in the Cu(Mn) case, only a slight tendency toward saturation was observed above about 80 kOe. In the Cu(Fe) system, no tendency whatsoever toward saturation was observed, even in fields up to 200 kOe. Gärtner *et al.*<sup>20</sup> also observed behavior in the Cu-Ni(Fe) alloy system which is qualitatively similar to our results in that the impurity contribution to the magnetoresistance scaled with Fe concentration and showed no tendency to saturate in fields up to 85 kOe.

Several suggestions have been advanced to ex-

plain the nonsaturation anomalies, including the one by Rohrer<sup>28</sup> and Liu<sup>30</sup> that the *s-d* model may simply be inadequate to account for the behavior of dilute magnetic alloys in a magnetic field. Rohrer<sup>8</sup> also indicates that some of the difficulty may be related to changes of the magnetic energy levels induced by the strong magnetic fields. Finally, it should be pointed out that the present theories do not take into account the influence of other scattering mechanisms such as phonons, other impurities besides the magnetic ones, and, in our case, the Ni on the magnetoresistance. In any event, it is significant that the magnetoresistance of the Cu-Ni (Mn) alloys discussed in this paper is negative, and this fact can be interpreted as support for the view that the Cu-Ni(Mn) samples are dilute magnetic alloys.

#### ACKNOWLEDGMENTS

The authors wish to acknowledge illuminating discussions with H. Gärtner and S. H. Liu. The assistance of C. Eagen in the resistivity measurements and of A. Johnson, H. Baker, F. A. Schmidt, and L. Reed in preparing the alloys is greatly appreciated.

\*Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission, Contribution No. 3068.

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