

University, Stanford, Calif. 94305.

¹K. Andres and E. Bucher, *Phys. Rev. Letters* **21**, 1221 (1968); **22**, 600 (1969); **24**, 1181 (1970), and references cited therein.

²B. Bleaney, *Proc. Roy. Soc. (London)* **276A**, 19 (1963); G. T. Trammell, *J. Appl. Phys.* **31**, 362S (1960); *Phys. Rev.* **131**, 932 (1963).

³Y.-L. Wang and B. R. Cooper, *Phys. Rev.* **172**, 539 (1968); **185**, 696 (1969).

⁴J. B. Gruber, E. A. Karlow, D. N. Olsen, and U. Ranon, *Phys. Rev. B* **2**, 49 (1970).

⁵T. E. Katila, E. R. Seidel, G. Wortmann, and R. L. Mössbauer, *Solid State Commun.* **8**, 1025 (1970).

⁶Other Tm salts have been investigated by this technique to 0.05 K, but no other examples of magnetic ordering have been found [T. E. Katila, V. K. Typpi, and E. R. Seidel, in *Proceedings of the Twelfth International Conference on Low Temperature Physics*, edited by E. Kanda (Keigaku, Tokyo, Japan, 1971), p. 711].

⁷B. B. Triplett, Ph.D. thesis (University of California, Berkeley, 1970) (unpublished).

⁸J. S. Griffith, *Phys. Rev.* **132**, 316 (1963).

PHYSICAL REVIEW B

VOLUME 6, NUMBER 5

1 SEPTEMBER 1972

Resistivity and Magnetoresistance of Dilute Solutions of Cr in Cu-Ni Alloys

C. F. Eagen and S. Legvold

*Ames Laboratory-USAEC and Department of Physics,
Iowa State University, Ames, Iowa 50010*

(Received 9 March 1972)

The electrical resistivity from 1.3 to 100 K and the longitudinal magnetoresistance from 0 to 85 kOe at 4.2 K were measured on Cu-Ni (Cr) alloys with Ni concentrations of 0, 6, 13, and 23 at. % and Cr concentrations of 0, 125, 300, 600, and 1200 at. ppm. All Cr-bearing samples were observed to exhibit resistivity minima. The difference in resistivity between each Cu-Ni (Cr) alloy and its Cr-free equivalent did not depend linearly on $\log_{10} T$; the difference in magnetoresistance between these two alloys was found to be negative. The Cr impurity contribution to the resistivity and magnetoresistance showed a marked dependence on the Ni concentration and was proportional to the Cr concentration only in the alloys containing 23 at. % Ni. These results are discussed in terms of spin-flip-scattering processes, characteristic of the Kondo effect, subject to Cr-Cr interactions and the local Ni environment around a Cr cell.

I. INTRODUCTION

The first major breakthrough in the understanding of the resistance minimum exhibited by dilute magnetic alloys was Kondo's¹ paper. He assumed that localized magnetic moments resided on the impurity sites and would interact with the conduction electron via the *s-d* Hamiltonian. The success and the shortcomings of this second-Born-approximation calculation, along with the works of Daniel and Friedel² and Anderson³ on the description of localized magnetic states in metals, have stimulated much research in recent years. Excellent review articles covering the progress made up to the late 1960's have been given by Daybell and Steyert,⁴ Kondo,⁵ and Heeger.⁶ A more recent theoretical review has been provided by Fischer.⁷

The formation and magnitude of the local moment depends on the delicate interplay of three quantities: (i) the position of the Fermi level relative to the energy of the *d*-state resonance; (ii) the density of states of the host metal at the energy of the *d*-state resonance; and (iii) the strength of the exchange and correlation effects (these are responsible for Hund's rules for atoms)

which cause the spin splitting of the virtual bound state. It is well established that Fe, Mn, and Cr all exhibit local-moment behavior in Cu,⁴ as well as resistivity minima.⁸⁻¹⁰ In addition, all three systems show a negative magnetoresistance.^{8,10,11} Thus, this set of alloys provides an excellent opportunity to study how the altering of the Cu host in some continuous fashion affects the resistivity and magnetoresistance of these alloys.

The first work in this direction was done by Gärtner *et al.*^{12,13} on Cu-Ni(Fe) alloys with Ni concentrations of 6, 12, and 23 at. % and Fe concentrations up to 1100 at. ppm. For fixed Ni concentrations, they found that the impurity contribution to the resistivity was proportional to the Fe concentration. However, the slopes of the $\log_{10} T$ plots of the resistivity decreased with increasing Ni concentration. Concurrent work by Bennett *et al.*¹⁴ on the Mössbauer effect in Cu-Ni(Fe) alloys showed evidence for the existence of magnetic Fe-Ni clusters.

Harvey *et al.*¹⁵ continued the investigation by measuring the resistivity and magnetoresistance of Cu-Ni (Mn) alloys with concentrations comparable to those in Gärtner's study. They found that the impurity contributions to the resistivity

and magnetoresistance were proportional to the Mn concentration and were essentially independent of the Ni concentration.

The present paper is concerned with the effect of alloying Ni into the Cu-Cr system. Concentrations were chosen so that this study would be comparable to the previous two. However, a strong Cr-Cr interaction adds an extra complication to this study that was not present in the previous investigations.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

The Cu-Ni master alloys were prepared from 99.999%-pure Cu and 99.999%-pure Ni obtained from the American Smelting and Refining Co. and the Atomerig Chemetals Co., respectively. The largest magnetic impurity present in either starting material was Fe, which was less than 0.7 wt ppm in the Cu and 3 wt ppm in the Ni. The base metals were first electron-beam melted under high vacuum in order to remove volatile impurities and then arc melted together in a shallow graphite crucible to form three alloys of 6, 13, and 23 at. % Ni. The graphite crucible was necessitated by the high melting point of Ni relative to Cu. Crystal bar Cr of 99.99+% purity, obtained from the Chromalloy Corp., was arc melted with Cu to form a 1-at. %-Cr master alloy. This alloy was then arc melted in appropriate quantities with the aforementioned Cu-Ni alloys to obtain Cu-Ni(Cr) alloys with nominal Cr concentrations of 125, 300, 600, and 1200 at. ppm.

The resulting fingers were swaged and drawn through a tungsten carbide die into wires of approximately a 0.04-in. diameter. Two samples, about 1 in. long, were cut from the parent wire and electropolished in a solution of three parts methanol to one part nitric acid. The samples were sealed in evacuated quartz ampoules and annealed for 3 days at 1000 °C. At the end of the annealing process the samples were quenched as rapidly as possible in ice water. About a 6-in. piece of the parent wire was used for analysis. The results are given in Table I.

In recent years there has been a considerable amount of work done on the magnetic properties of Cu-Ni alloys. For Ni concentrations in excess of 47 at. %, the alloys are ferromagnetic, while for Ni concentrations between 30 and 47 at. %, giant magnetic-polarization clouds tend to be associated with the Ni-rich regions. It is expected that such regions will occur on a purely statistical random basis and, conceivably, through sample processing. According to Houghton *et al.*,¹⁶ the spin-flip scattering of the conduction electrons with these clouds is responsible for the

maxima and minima observed in the resistivity and the negative magnetoresistance of these alloys. Seib and Spicer¹⁷ have given a fairly extensive discussion on this cluster phenomenon. They conclude that with the annealing process described above for the present work, and for Ni concentrations less than 23 at. %, there should be little or no departure from randomness in the Cu-Ni alloy system, and little or no cluster-scattering effects should be present.

The resistivity and magnetoresistance measurements were made by the standard dc four-probe technique using a potentiometer with a resolution of ± 5 nV and a photocell amplifier. The currents employed were less than 200 mA and stable to better than one part in 10^5 . Au-Fe and constantan-vs-Cu thermocouples were used to determine the temperature, and magnetic fields were supplied by a 100-kOe superconducting solenoid.

An uncertainty of about 10% in the Cr concentrations in our samples made estimates of errors due to apparatus effects relatively unimportant. The ubiquitous problem of He absorption on the surface of the sample is believed to account for the slight cusps at 4.2 K evident in some of the $\log_{10} T$ plots of the resistivity. As a check on systematic errors and solubility problems, the second sample, for a few concentrations, was measured. In no cases were there any significant deviations in the data between the two samples. No studies were made on the possible effects of different annealing times and temperatures.

TABLE I. Analysis of the samples; the value of the temperature at which the minimum occurs, T_{\min} ; the impurity contribution to the resistivity at 1.3 K minus the value at T_{\min} ; the residual resistivity; and the impurity contribution to the magnetoresistance at 85 kOe.

Sample (at. % Ni) (at. ppm Cr)	T_{\min} (K)	$\Delta\rho(1.3\text{ K})-\Delta\rho(T_{\min})$ ppm (nΩ cm)	$\rho(4.2\text{ K})$ (μΩ cm)	$\Delta\rho(85\text{ kOe})$ ppm (nΩ cm)
Cu	0.010	...
Cu-Cr(114)	23.0	0.404	0.149	-0.070
Cu-Cr(286)	26.0	0.323	0.319	-0.076
Cu-Cr(645)	28.5	0.178	0.493	-0.053
Cu-Cr(1236)	30.5	0.103	0.789	-0.037
Cu-5.5Ni	5.98	...
Cu-5.5Ni-Cr(138)	21.5	0.342	6.11	-0.102
Cu-5.5Ni-Cr(301)	26.0	0.345	6.23	-0.097
Cu-5.4Ni-Cr(645)	30.0	0.250	6.40	-0.065
Cu-5.5Ni-Cr(1185)	34.5	0.216	6.95	-0.054
Cu-12.8Ni	14.0	...
Cu-12.6Ni-Cr(160)	22.0	0.200	14.0	-0.050
Cu-12.7Ni-Cr(278)	25.5	0.201	14.0	-0.049
Cu-12.6Ni-Cr(531)	27.0	0.166	14.0	-0.043
Cu-12.4Ni-Cr(1205)	34.5	0.164	14.0	-0.035
Cu-22.9Ni	25.3	...
Cu-22.9Ni-Cr(140)	19.0	0.055	25.3	-0.024
Cu-22.9Ni-Cr(327)	23.0	0.066	25.3	-0.016
Cu-23.0Ni-Cr(618)	25.0	0.072	25.3	-0.017
Cu-22.6Ni-Cr(1209)	30.0	0.080	25.3	-0.017

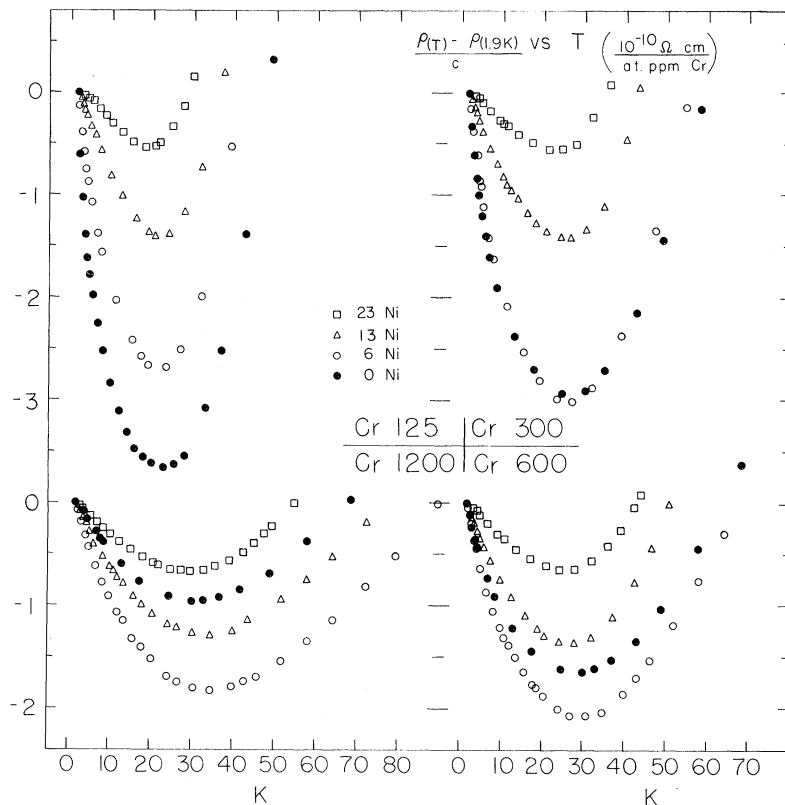


FIG. 1. Resistivity minus the value of the resistivity at 1.9 K.

III. RESULTS

The electrical resistivity of the alloys listed in Table I was measured as a function of temperature from 1.3 to 100 K. The resistivity data for the Cr-bearing samples are shown in Fig. 1, where the curves have been divided by the Cr concentrations to facilitate comparison. All the Cr-bearing samples exhibit resistivity minima, and below the temperature at which the minimum occurs, T_{\min} , no evidence was found for a resistivity maximum. The resistivities of the host alloys are shown in Fig. 2. The appropriate values for the residual resistivities can be found in Table I. The impurity contribution to the resistivity per at. ppm of Cr is plotted as a function of $\log_{10} T$ in Figs. 3 and 4. This quantity is defined as the difference in the resistivity between a Cr-bearing alloy and its Cr-free equivalent. The values of this quantity at 1.3 K minus the values at T_{\min} are listed in Table I. These values, rather than the more traditional depth of the minimum, are used as a measure of the strength of the spin-flip scattering. Values of the residual resistivity $\rho(4.2 \text{ K})$ are given in Table I. It was necessary to average the residual values of the 13- and 23-at.-%-Ni samples over the various Cr concentrations due to the errors in determining their respective geometric factors. By comparing our data for

a Cu-Cr (114-at. ppm-Cr) sample between 1 and 4 K with a Cu-Cr (113-at. ppm-Cr) sample as measured by Daybell and Steyert,¹⁰ it was found that our data would seem to provide a smooth extension of theirs into the region of higher Cr concentrations.

The longitudinal magnetoresistance at 4.2 K was measured as a function of field from 0 to 85 kOe. Figures 5-8 show the magnetoresistance $\rho(H) - \rho(0)$ for fixed Ni concentration. The magnetoresistance of the Cr-free samples behaves in the "normal" fashion in that it increases with increasing magnetic field. The magnetoresistance of a Cr-bearing sample minus the magnetoresistance of its Cr-free equivalent, which we denote by $\Delta\rho(H)$, is negative. Values of this quantity per at. ppm of Cr evaluated at 85 kOe are given in Table I.

In Fig. 3 we see that the curves for the lowest Ni concentrations are not proportional to the Cr concentration. This behavior, and the deviation from linearity of the $\log_{10} T$ plots for the higher concentration Cu-Cr alloys, will be discussed in terms of Cr-Cr interaction effects. The fact that the resistivities of the Cu-23-at.-%-Ni (Cr) alloys, shown in Fig. 3, are proportional to the Cr concentration will be discussed in terms of the extra Coulomb scattering afforded by the Ni impurities and its effect on the mean free path of the conduc-

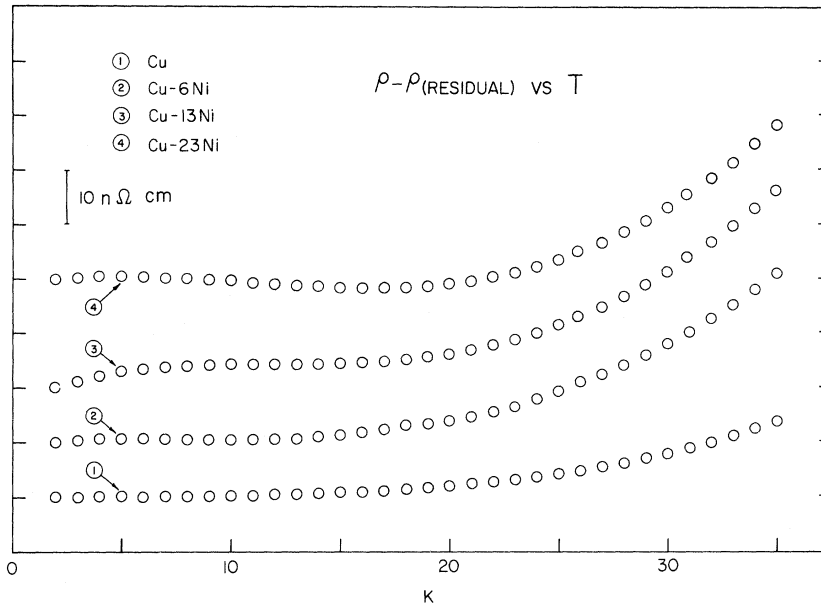


FIG. 2. Resistivity of the host alloys.

tion electrons. Finally, the extreme departure from linearity and the reduction in size of the impurity contributions to the resistivity of the higher-Ni-concentration alloys will be interpreted in terms of local Ni-Cr interaction effects. Since the theory of the magnetoresistance of dilute magnetic alloys and its separation into component parts is not as well understood as the resistivity, the magnetoresistance data will be used only as a check on the validity of the hypothesis put forward to explain the resistivity data.

IV. DISCUSSION

A. Resistivity

The early theory of dilute magnetic alloys given by Kondo¹ predicts that the spin-flip-scattering contribution to the resistivity $\Delta\rho$ should be given by

$$\Delta\rho(T) = cR_u (\pi\rho_1 J)^2 S(S+1) [1 + 4\rho_1 J \ln(T/T_0)] . \quad (1)$$

In this expression c is the atomic concentration of magnetic impurities, R_u is the unitarity limit of the resistivity which is a constant for a given host, ρ_1 is the density of the conduction-electron states at the Fermi level for one direction of the spin, J is the strength of the s - d exchange interaction which is negative, S is the spin on the impurity, and T_0 is a characteristic temperature. This expression is restricted to temperatures greater than a characteristic temperature that signals the breakdown of perturbation theory and the onset of a many-body singlet state. In deriving Eq. (1) it is tacitly assumed that the impurity concentrations are so dilute that the con-

tribution to the resistivity by each impurity is not influenced by the presence of the other impurities.

When this assumption is not satisfied, the problem becomes an extremely difficult one. One can get a qualitative idea of the behavior of such an alloy by examining the somewhat idealized problem of two interacting magnetic impurities in a conduction sea. In attacking this problem one assumes that the additional term to the Hamiltonian takes the form

$$H_w = -W(R) \vec{S}_1 \cdot \vec{S}_2 , \quad (2)$$

where \vec{S}_1 is the spin on one impurity, \vec{S}_2 is the spin on the second impurity which is separated from the first by a distance R , and $W(R)$ is the coupling energy. Heuristically, we see that if we replace $W(R) \vec{S}_2$ by an effective magnetic field \vec{H}_1 , then the pair-impurity problem can be thought of in terms of the magnetoresistance problem for one impurity.

A quantitative understanding of the magnetoresistance problem has proved to be elusive. A qualitative understanding may, however, be obtained as follows: In zero applied field it is usually assumed that the impurity-spin components M_s are degenerate. Thus, in the first Born approximation the conduction electrons suffer an elastic spin-flip scattering with the impurity spin. In the presence of a magnetic field the conduction band and the impurity levels are Zeeman split. The preferential aligning of the impurity spins causes the spin-flip-scattering process to become anisotropic and inelastic. For values of $\mu_B H$ large compared to $k_B T$, where

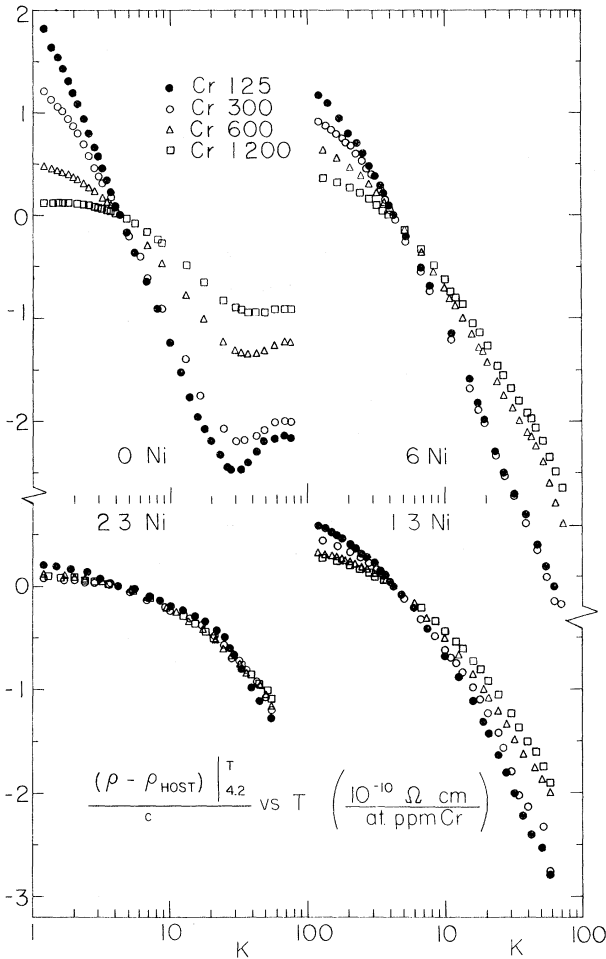


FIG. 3. Impurity contribution to the resistivity minus its value at 4.2 K.

μ_B is the Bohr magneton and k_B is Boltzmann's constant, it becomes increasingly difficult for an electron within $k_B T$ of the Fermi level to find an unoccupied state a distance $2\mu_B H \pm k_B T$ below the Fermi level. Hence, a magnetic field tends to suppress the spin-flip scattering that is responsible for the rise in the resistivity at temperatures below the minimum temperature.

Matho and Béal-Monod¹⁸ have investigated the pair-impurity problem in some detail. In the limit of large values of βW and ferromagnetic coupling ($W > 0$), they find the spin contribution to the resistivity can be expressed in the tractable form

$$\Delta\rho(\beta W) = cR_u (\pi\rho_1 J)^2 S_{eff}^2 (\beta W) [1 + 4J\rho_1 \ln(T/T_0) + 2J\rho_1 \ln(1 + T_W^2/T^2)^{1/2}], \quad (3)$$

with

$$\beta = 1/k_B T; \quad T_W = (2SW/k_B) (T_0/T_F), \quad (4)$$

where T_F is the Fermi temperature. There are explicit and implicit concentration dependences in Eq. (3). The explicit dependence c suggests that one may simply sum the contributions to the resistivity from isolated impurity pairs. Next it may be noted that W is a function of R ; therefore, it is necessary to use some sort of a mean value for T_W and S_{eff} that will be concentration dependent. It is also interesting to note that in this limit we can think of the resistivity as being the sum of two terms. The first term is the simple Kondo expression with a modified magnetic moment, which tends to saturate for large values of βW , and the second term is only appreciable for temperatures less than T_W .

In Fig. 3 we see that the resistivity curves of the Cu-Cr alloys do exhibit behavior in qualitative agreement with this interaction interpretation. That is, $\Delta\rho$ exhibits a definite nonlinear behavior with $\log_{10} T$ which becomes more pronounced as the Cr concentration is increased. Daybell and Steyert¹⁰ have observed these interaction effects for Cr concentrations as low as 50 at. ppm. By proceeding clockwise around Fig. 3, we see how the addition of Ni affects the impurity contribution to the resistivity. The most striking result

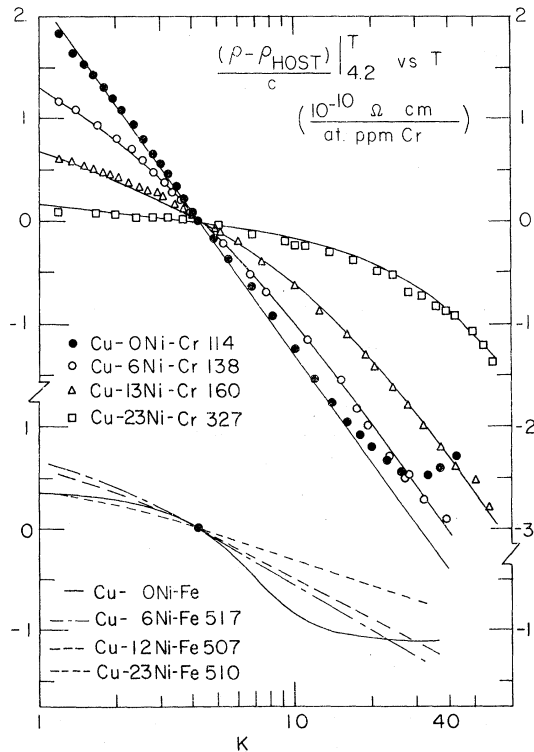


FIG. 4. Impurity contribution to the resistivity, the fit of Eq. (6) to the data, the data of Gärtner *et al.* (Ref. 12) for Cu-Ni (Fe), and the data of Daybell and Steyert (Ref. 9) for Cu-Fe.

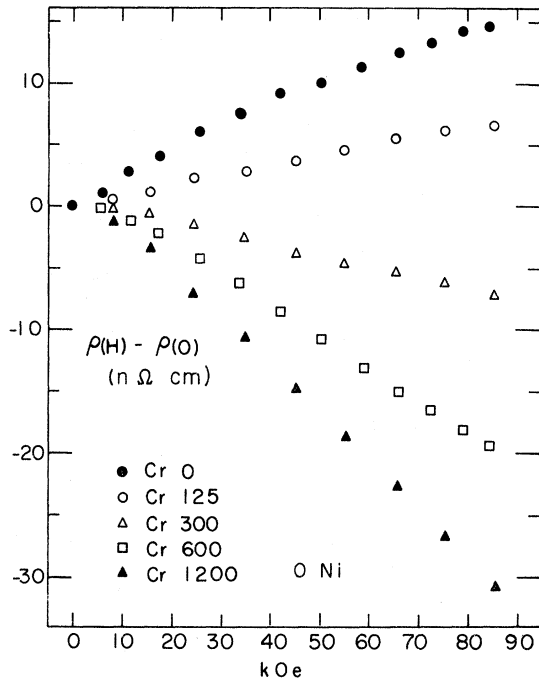


FIG. 5. Magnetoresistance of the Cu-Cr alloys.

is that for the 23-at.-%-Ni samples the resistivity is proportional to the Cr concentration. We believe that this is due to the extra Coulomb scattering afforded by the Ni impurities. The Cr-Cr interaction, which is responsible for the departure from proportionality, must be very long range. The scattering from the intervening Ni sites will decrease the mean free path of the conduction electrons and, thereby, decrease the range of the Rudermann-Kittel-Kasuya-Yosida (RKKY) interaction. Therefore, we feel that in the samples with low Cr and high Ni concentrations the scattering processes of interest can be characterized by a Cr ion and its local Cu-Ni environment. We notice that the $\log_{10} T$ plots of the 23-at.-%-Ni samples show a large deviation from linearity. In an attempt to glean some information from the data about this effect, we have plotted the resistivity of the lower Cr concentrations for the various Ni concentrations, which is shown in Fig. 4. Although the Cr-Cr interactions are not negligible for the lower Ni concentrations shown in Fig. 4, we do expect the predominant effect on the resistivity to be due to the local Ni environment around a Cr cell.

Trying to deduce from macroscopic measurements, such as the resistivity, how the alloying of Ni affects the basic spin-flip scattering is not an easy task. However, there are known facts about similar systems which, coupled with some speculation, may open up some useful avenues

for thought about this system. Seib and Spicer¹⁷ have found that the virtual-bound-state model seems to be the appropriate one in describing Cu-Ni alloys. They estimate that the Ni d state has a resonance at about 0.95 eV below the Fermi level with a level width of at least 0.8 eV. The work of Harvey *et al.*¹⁵ on Cu-Ni (Mn) alloys, at temperatures and Mn concentrations where Mn-Mn interactions were negligible showed that the Ni had little effect on the resistivity. This is apparently related to the fact that the Mn d states are strongly spin split and far away in energy from the Ni d -state resonances. Thus, the Ni has a small effect on the local-moment formation at a Mn site and, together with the small value of J for Cu-Mn, puts any alloying effects below the resolution of the resistivity measurements made.

The Ni in Cu-Ni (Fe) alloys has a pronounced effect, as referenced in Sec. I. The d states in Fe are not as strongly spin split as they are in Mn, and the Ni d -state and Fe d -state resonances may overlap in energy. The result is an enhanced magnetic moment in the region of the Fe impurity. The size of this moment has been estimated¹⁴ to be

$$\mu = (2.85 + 0.6n)\mu_B, \quad (5)$$

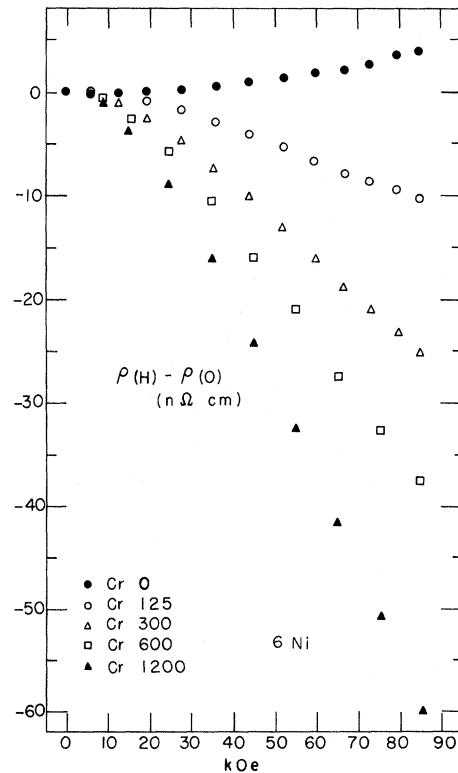


FIG. 6. Magnetoresistance of the Cu-6-at.-%-Ni(Cr) alloys.

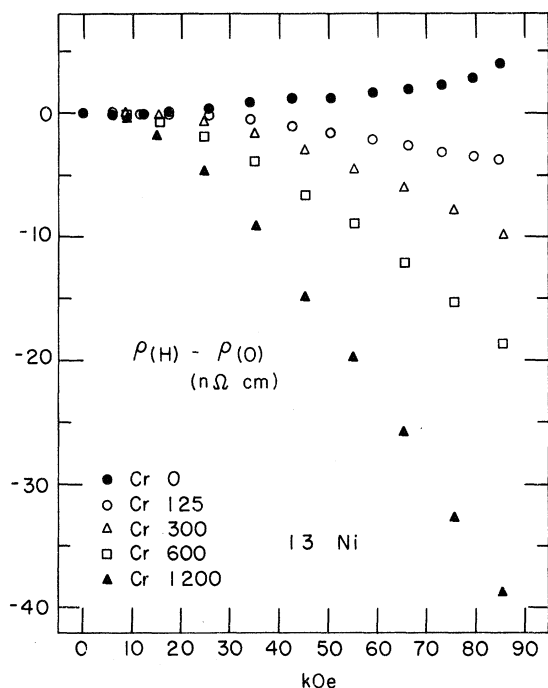


FIG. 7. Magnetoresistance of the Cu-13-at.-%-Ni(Cr) alloys.

where n is the average number of Ni nearest neighbors expected at an Fe site on a statistical basis. The resistivity of these alloys is plotted in Fig. 4. We notice that for temperatures above 4 K the curves have a $\log_{10} T$ dependence with a slope that is a function of the Ni concentration. This leads one to speculate that the total moment undergoes elastic spin-flip scattering with values of J and S that depend on the number of Ni nearest neighbors at the Fe site.

In some ways the properties of the Cu-Ni (Cr) alloys seem to lie halfway between those of the Cu-Ni (Fe) and Cu-Ni (Mn) systems. The local Ni environment has a pronounced effect, but at temperatures above 20 K the slopes of the curves seem to asymptotically approach a constant value which is independent of the Ni concentration. (We put some reservations on this statement in regard to the 23-at.-%-Ni alloys.) In addition, the temperature at which the minimum occurs is relatively insensitive to the Ni concentration. This leads us to conclude that in Cu-Ni (Cr) alloys the appropriate picture is one where the low-temperature resistivity is suppressed, but above a certain temperature the resistivity is essentially independent of the Ni concentration.

Comparison of these properties to those of Eq. (3) led us to try to fit the data by the expression

$$\Delta\rho = -c(A - B) \log_{10} T - cB \log_{10} (T^2 + \theta^2)^{1/2} + \rho_0, \quad (6)$$

where B is concentration dependent and ρ_0 is related to the residual resistivity. The value of A was determined by the slope of the Cu-Cr (114) sample. The results of this handfit are represented in Fig. 4 by the solid curves, and the corresponding values of A , B , and θ are given in Table II. The fact that we obtain a good fit to the data with this functional form is interesting, but it appears that any significance in the values of A , B , and θ is masked by the Cr-Cr interaction effects in the data.

It is tempting to interpret our data in terms of magnetic clusters similar to those seen in Cu-Ni (Fe) alloys. However, this seems to be too stringent a postulate in the case of Cu-Ni (Cr). In the case of Cu-Ni (Fe) the moment of the cluster seems to flip as a rigid paramagnetic unit. This implies elastic scattering which cannot account for the suppression effect seen in Cu-Ni (Cr). Gainon and Heeger¹⁹ have studied Cu-Mn alloys doped with dilute amounts of Pt. They observe a suppression of the resistivity at low temperatures which closely resembles the suppression in our alloys. They attribute this suppression to spin-orbit interactions. This mechanism should only be appreciable for large Z impurities, such as Pt, and should be negligible for the case of Ni in Cu.

Riess and Ron²⁰ have studied the suppression of the resistivity due to inelastic scattering of

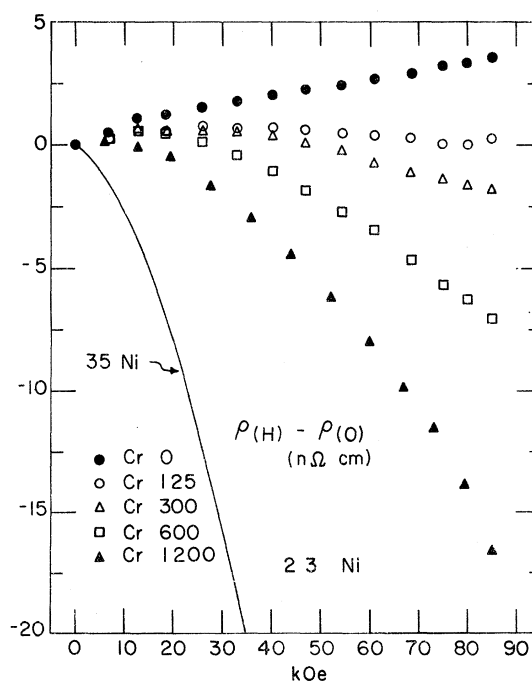


FIG. 8. Magnetoresistance of the Cu-23-at.-%-Ni(Cr) alloys. (Note the change of scale in this figure.)

TABLE II. Values of A , B , and θ defined in Eq. (6) and the actual values of $\Delta\rho/\text{ppm}$.

Sample (at. % Ni) (at. ppm Cr)	A (n Ω cm)	B (n Ω cm)	θ (K)	$\Delta\rho(1.2\text{ K})$ ppm (n Ω cm)
Cu-Cr(114)	0.3452	0.0000	...	1.4
Cu-5.5Ni-Cr(138)	0.3452	0.1806	4.51	1.1
Cu-12.6Ni-Cr(160)	0.3452	0.2450	11.51	0.9
Cu-22.9Ni-Cr(327)	0.3452	0.3156	34.75	4.0

the conduction electrons. They treat interaction effects, such as the RKKY interaction between two impurities, in terms of a broadening of the width of the virtual bound state. Although their study is primarily concerned with the suppression of the resistivity below the maximum that often occurs in dilute magnetic alloys, they do show that this broadening introduces inelastic-scattering processes. This might be a fruitful approach in understanding Cu-Ni(Cr) alloys. For our purposes, it seems to be convenient and sufficient to treat the interaction between the Ni d states and the Cr d state by a local internal magnetic field \vec{H}_1 produced by the Ni at the Cr site. If we picture the magnetic moment as residing in the Cr cell under the influence of this local field, whose strength depends on the number of Ni nearest neighbors, then we have a mechanism for inelastic scattering that is independent of the Cr concentration. This picture is further motivated by the similarity of Fig. 4 to the magnetoresistance curves of Daybell and Steyert¹⁰ for a 28 at. ppm Cr in Cu sample.

By constraining our data at 4.2 K, we have suppressed some important information. In Table II we show the actual values of $\Delta\rho/\text{ppm}$ at 1.2 K. Up through 13 at. % Ni the values are not surprising, but the 23-at. %-Ni sample shows a marked anomaly. This could signify that at these Ni concentrations we are no longer dealing with the influence of just a few Ni ions around a Cr site.

B. Magnetoresistance

Figures 5–8 show the longitudinal magnetoresistance at 4.2 K for the various Ni concentrations. The extra complications due to the Cr-Cr interactions, along with the inherent difficulty of separating the “normal” positive contribution to the magnetoresistance from the negative contribution, legislate to make a detailed analysis of the data marginal. A fairly complete summary of the influence of a magnetic field on the resistivity can be found in the paper by Harvey *et al.*¹⁵ and the references cited therein.

If the interpretation in Sec. III of the effect of the local Ni environment on the spin-flip scattering is right, then the magnetoresistance should be

a function of two magnetic fields. Béal-Monod and Weiner²¹ indicate that if the population of the Zeeman levels is governed by the Boltzmann factor, then the spin-flip-scattering contribution to the magnetoresistance should be “frozen out” by fields which satisfy the relation

$$g\mu_B H/k_B T > 4. \quad (7)$$

Setting $g = 2$ and $T = 4.2$ K, expression (7) implies that saturation should tend to occur for $H > 125$ kOe. For the following discussion we will be interested in the values of $\Delta\rho(85)/\text{ppm}$ which appear in Table I.

The fact that this quantity is negative is support for the view that we are still dealing with a magnetic alloy. For the higher Ni concentrations it appears that $\Delta\rho(85)/\text{ppm}$ may be proportional to the Cr concentration. This trait can be given the same interpretation that was given for the resistivity. If we look at the same set of samples that were analyzed in Fig. 4, we see that $\Delta\rho(85)/\text{ppm}$ first “increases” for the addition of 6 at. % Ni and then steadily “decreases” as the Ni concentration is further increased. We feel that this behavior is consistent with the picture of an internal magnetic field.

For 114 at. ppm Cr in Cu, one has an effective internal field due to the Cr-Cr interactions. This field has already frozen out much of the spin-flip scattering. The effect of the external field will, therefore, be less in the presence of this internal field. As we increase the Cr concentration in the Ni-free alloys, we see that $\Delta\rho(85)/\text{ppm}$ tends to decrease further in agreement with this interpretation. As was mentioned earlier, the alloying of Ni tends to do two things: It introduces extra scattering that decreases the strength of the Cr-Cr interactions, and it also produces an internal field at the Cr site. In the 6-at. %-Ni alloys there is a 50% chance that a Cr ion will not have any Ni ions as nearest neighbors. It therefore seems plausible to assume that the former effect will be the predominant one in the 6-at. %-Ni alloys. This will decrease the strength of the internal field with respect to the Ni-free alloys, and a larger amount of the freezing will now have to be done by the external field. This will cause $\Delta\rho(85)/\text{ppm}$ to increase. If upon the alloying of more Ni the second effect becomes predominant, then \vec{H}_1 should increase with a corresponding lowering of the values of $\Delta\rho(85)/\text{ppm}$. Thus, we conclude that the magnetoresistance data are consistent with the interpretation given to the resistivity data.

One is tempted to argue on the basis of expression (7) that the samples with a large value for \vec{H}_1 should tend to saturate faster than those with small \vec{H}_1 . Cu-Cr(114) does show strong saturation effects; Cu-23-at. %-Ni-Cr(327) looks as if

it is saturating, and the other two samples are definitely not saturating at 85 kOe. However, one must not take these results too seriously. From the experimental standpoint, the prediction of saturation is very seldom realized in dilute magnetic alloys. Some authors attribute this to the inadequacy of perturbation theory, and others^{22, 23} feel that the s - d model may simply be inadequate in the presence of a magnetic field.

ACKNOWLEDGMENTS

We would like to thank D. T. Peterson, who supervised the sample preparation, and A. R. Harvey for his stimulating and pointed discussions. We also wish to acknowledge the various contributions made by S. H. Liu, A. Johnson, H. Baker, F. A. Schmidt, L. Reed, T. Plant, M. Pitchford, J. Barnes, and P. Dishaw.

- ¹J. Kondo, *Progr. Theoret. Phys. (Kyoto)* **32**, 37 (1964).
- ²E. Daniel and J. Friedel, in *Proceedings of the Ninth International Conference on Low Temperature Physics, Columbus, Ohio, 1964*, edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum, New York, 1965).
- ³P. W. Anderson, *Phys. Rev.* **124**, 41 (1961).
- ⁴M. D. Daybell and W. A. Steyert, *Rev. Mod. Phys.* **40**, 38 (1968).
- ⁵J. Kondo, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1968), Vol. 23.
- ⁶A. J. Heeger, in Ref. 5, Vol. 23.
- ⁷K. Fischer, *Phys. Status Solidi* **46**, 11 (1971).
- ⁸P. Monod, *Phys. Rev. Letters* **19**, 1113 (1967).
- ⁹M. D. Daybell and W. A. Steyert, *Phys. Rev. Letters* **18**, 398 (1967).
- ¹⁰M. D. Daybell and W. A. Steyert, *Phys. Rev. Letters* **20**, 195 (1968).
- ¹¹H. Rohrer, *J. Appl. Phys.* **40**, 1472 (1969).
- ¹²H. Gärtner, D. R. Zrudsky, and S. Legvold, *Solid State Commun.* **8**, 913 (1970).
- ¹³H. Gärtner, A. R. Harvey, D. R. Zrudsky, and S. Legvold, *Solid State Commun.* **8**, 1975 (1970).
- ¹⁴L. H. Bennett, L. J. Swartzendruber, and R. E. Watson, *Phys. Rev. Letters* **23**, 1171 (1969).
- ¹⁵A. R. Harvey, S. Legvold, and D. T. Peterson, *Phys. Rev. B* **4**, 4003 (1971).
- ¹⁶R. W. Houghton, M. P. Sarachik, and J. S. Kouvel, *Solid State Commun.* **8**, 943 (1970).
- ¹⁷D. H. Seib and W. E. Spicer, *Phys. Rev. B* **2**, 1676 (1970).
- ¹⁸K. Matho and M. T. Béal-Monod, *Phys. Rev. B* **5**, 1899 (1972).
- ¹⁹D. Gainon and A. J. Heeger, *Phys. Rev. Letters* **22**, 1420 (1969).
- ²⁰I. Riess and A. Ron, *Phys. Rev. B* **4**, 4099 (1971).
- ²¹M. T. Béal-Monod and R. A. Weiner, *Phys. Rev.* **170**, 552 (1968).
- ²²H. Rohrer, *Phys. Rev.* **174**, 583 (1968).
- ²³S. H. Liu (private communication).