

## Magnetic field depreciation of the Kondo resistivity in $\text{Cu}_{1-X}\text{Au}_X - \text{Cr}$

E. W. Fenton

National Research Council of Canada, Ottawa, Canada

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The addition of a few atomic percent of gold to the copper-chromium Kondo system acts to suppress the normal positive magnetoresistance to negligible levels, for magnetic fields up to 100 kG. The negative component of the magnetoresistance due to field depreciation of the Kondo state has been determined. No peak occurs in curves of Kondo resistivity vs temperature in the presence of magnetic fields up to 100 kG, but instead the Kondo resistivity saturates at a low-temperature plateau in high fields.

In two recent papers I discussed measurements of resistivity in  $\text{Cu}_{1-X}\text{Au}_X - \text{Fe}$  and  $\text{Cu}_{1-X}\text{Au}_X - \text{Cr}$  systems (with  $X$  a few atomic percent) in the presence of magnetic fields as large as 100 kG, with the current parallel to the field.<sup>1</sup> The addition of the third gold constituent to the well-studied Cu-Fe and Cu-Cr systems was designed to achieve two objectives: (a) deviations from Matthiessen's rule, when separating the electron-phonon part of the resistivity, would be moved to higher temperatures; (b) the normal magnetoresistance would be reduced to a negligible level. The technique of adding a third constituent to obtain magnetoresistance measurements with effects dominated by the Kondo state was successful for the  $\text{Cu}_{1-X}\text{Au}_X - \text{Fe}$  systems, but not for the  $\text{Cu}_{1-X}\text{Au}_X - \text{Cr}$  systems. Attempts to dissolve Cr in  $\text{Cu}_{1-X}\text{Au}_X$  matrices were only partially successful. Cr is considerably less soluble than Fe in a Cu matrix. From our work Cr is apparently even less soluble in  $\text{Cu}_{1-X}\text{Au}_X$ , with  $X$  a few atomic percent. In the work discussed in Ref. 1, for a  $\text{Cu}_{99.5}\text{Au}_{0.5} - \text{Cr}_{0.0025}$  ingot approximately 80% of the nominal Cr concentration was shown in chemical analysis. The combined potential scattering (nonmagnetic) from Au and Cr atoms in this alloy was not sufficient to suppress the normal magnetoresistance to negligible values. For a nominal  $\text{Cu}_{98.25}\text{Au}_{1.75} - \text{Cr}_{0.0035}$  alloy, only about 15% of the Cr appeared in chemical analysis. For this alloy, resistivity measurements showed no Kondo-resistivity component at all, not even the component that would be expected for the 0.00055-at. % Cr concentration shown in the chemical analysis.

Recently  $\text{Cu}_{98.25}\text{Au}_{1.75} - \text{Cr}_{0.0035}$  alloys were prepared wherein chemical analysis shows the Cr present to be 0.0030 at. %. Variation less than 0.0003 at. % from this concentration occurred for eight regions of the ingot. (The chemical analysis was done spectrographically by carrier distillation, calibrated using wet-chemistry techniques.) The preparation of this alloy involved an improved technique with a considerably more violent quench from the liquid state.  $\text{Cu}_{98.25}\text{Au}_{1.75}$  and Cr were

heated in an induction furnace under vacuum, followed by quenching from the melt in cold water. The relatively concentrated Cr-matrix system that resulted was diluted with addition of  $\text{Cu}_{98.25}\text{Au}_{1.75}$  stock, again with a violent quench from the melt. The final alloy was annealed for three days at 950 °C in an evacuated and sealed quartz tube.

The longitudinal resistivity was measured as discussed in Ref. 1, with specimens in a <sup>3</sup>He cryostat inserted into the bore of a superconducting solenoid. In Fig. 1 the effects of temperature and magnetic field on  $\Delta\rho = [\rho(\text{Cu}_{98.25}\text{Au}_{1.75} - \text{Cr}_{0.0035}) - \rho(\text{Cu}_{98.25}\text{Au}_{1.75})]/c$  are shown.  $c$  is the Cr concentration. Measurements on two pairs of specimens, each pair with Kondo alloy and reference matrix, were the same to within deviations much less than the error-bar estimates shown. The error bars are due principally to uncertainties in specimen dimensions, about 0.5% for each pair. At room temperature the electron-phonon contribution to the resistivity is much larger than the  $\Delta\rho$  compo-

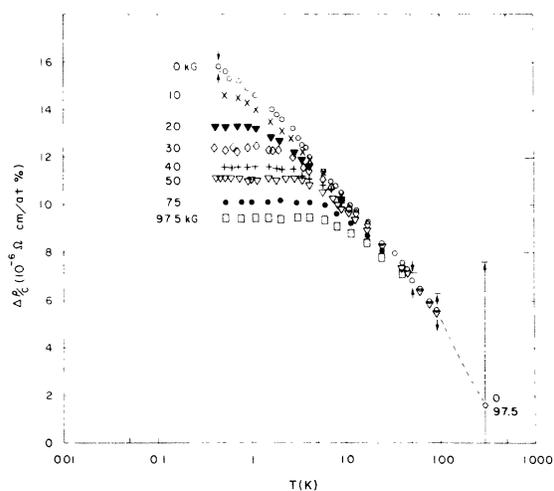


FIG. 1. Dependence of the function  $\Delta\rho_c = [\rho(\text{Cu}_{98.25}\text{Au}_{1.75}\text{Cr}) - \rho(\text{Cu}_{98.25}\text{Au}_{1.75})]/c$  on temperature and magnetic field.

ment, which means much greater uncertainty in  $\Delta\rho$ . Subtraction of the resistivity of the reference matrix from the Kondo alloy was accomplished by the direct-measurement procedure discussed in Ref. 1, but with voltage measurements for each specimen recorded as well, as a check against short circuits, etc. Measurements were made only for a single Cr concentration, which means that there was no check that  $\Delta\rho$  is linear in the concentration  $c$ . The 0.0035 at. % nominal and 0.0030 at. % analysis concentrations for Cr are both within the one-impurity linear range for the resistivity, shown in measurements on Cu-Cr by Daybell and Steyert.<sup>2</sup>

The combined effect of potential scattering from Au and Cr atoms in the  $\text{Cu}_{98.25}\text{Au}_{1.75}\text{-Cr}_{0.0035}$  system is sufficient to limit the normal longitudinal magnetoresistance, including a part from the Kondo resistivity, to a small fraction of 1%, or less, for fields to 100 kG. The magnetoresistance-limiting potential scattering is almost entirely due to the gold concentration which means that the magnetoresistance of the matrix, much less than 1%, almost entirely cancels itself in  $\Delta\rho$ . As discussed in Ref. 1, the normal positive magnetoresistance for the Kondo-resistivity term may be several times larger than it is for the normal potential-scattering part; however, this still means that the effects of normal magnetoresistance in Fig. 1 are less than 1% of  $\Delta\rho$ .

The resistivity results in Fig. 1 show magnetic-field depreciation of the Kondo state, in a similar manner to the effect of increased temperature. In Ref. 1, the theory of the Kondo resistivity in a magnetic field is reviewed, and we refer here as well to the work by More and Suhl<sup>3</sup> and by Bloomfield, Hecht, and Sievert.<sup>4</sup> The theory for  $H \neq 0$  is not as well developed or as reliable as for the  $H = 0$  case. Theories for the high-field perturbation regime and for a low-field anomalous Green's function regime suggest that magnetic field and temperature act on Kondo systems in a similar manner, with the field effects scaled according to an  $H_K$  given by

$$Sg\mu_B H_K = Ak_B T_K, \quad (1)$$

where  $A$  is a dimensionless constant which is expected to be near unity.

Theories which are essentially nonperturbative and do not use a ground-state *ansatz* for the ( $H = 0$ ,  $T = 0$ ) condition suggest that field effects will be qualitatively different from the behavior of  $\Delta\rho$  vs  $T$  at  $H = 0$ . One peak, or several peaks, are predicted in the  $\Delta\rho$  vs  $T$  curve for high fields.<sup>3,4</sup> Although impurity-impurity interactions have long been known to cause such peaks in the resistivity,<sup>5</sup> to our knowledge there is no evidence of this behavior for Kondo systems which are sufficiently dilute so that the resistivity is in the one-impurity

regime where  $\Delta\rho$  is linear in  $c$ . In particular, our own results for the resistivity in  $\text{Cu}_{1-x}\text{Au}_x\text{-Fe}$  alloys do not show this behavior<sup>1</sup>, nor do the results for Cu-Cr obtained by Daybell and Steyert.<sup>2</sup> The high-field results in Cu-Cr by Daybell and Steyert<sup>2</sup> are open to question due to their method for separating the normal positive magnetoresistance. However the results in Fig. 1, where this method is not a factor, support the qualitative result of Ref. 2, that no peaks occur in the curves  $\Delta\rho$  vs  $T$  at high fields. The results by Daybell and Steyert extended down to 0.04 K, whereas the results in Fig. 1 extend only to 0.4 K. However 0.4 K is obviously far below  $T_K$ , where the most prominent peak in  $\Delta\rho$  vs  $T$ , with  $H \neq 0$ , is predicted to occur.<sup>3</sup> Moreover, any temperature-dependent errors which might occur in the technique used by Daybell and Steyert for separating the normal positive magnetoresistance would be largest where the temperature dependence of the Kondo resistivity and/or the matrix resistivity is strongest. We can assume that the temperature dependence of  $\Delta\rho$  below 0.4 K in the results of Daybell and Steyert is valid, which means that the low-temperature plateaus in  $\Delta\rho$  vs  $T$  at high fields in Fig. 1 can be safely assumed to extend to the zero-temperature limit. The scaling temperature or energy in the Kondo system is  $k_B T_K$  and no new effects are expected at, say,  $T = 10^{-3} T_K$ . I cannot reconcile the experimental results in  $\text{Cu}_{1-x}\text{Au}_x\text{-Fe}$  or  $\text{Cu}_{1-x}\text{Au}_x\text{-Cr}$  systems with the predictions of the theories in Refs. 3 and 4 for  $H \neq 0$ . In the theory of Kondo resistivity for  $H = 0$ , early difficulties with spurious peaks occurred.<sup>6</sup> Although lessons have been learned from those difficulties, it is perhaps possible that further difficulties have been introduced in this regard by the  $H \neq 0$  condition, particularly in regard to analytic properties of the scattering matrix in the complex frequency plane.

In Fig. 1, and at roughly the half-height of the Kondo resistivity step, 97.5 kG is equivalent to 17 K in depreciating the Kondo resistivity. Assuming the  $g$  value is 2 and using  $S = \frac{3}{2}$  obtained in high-temperature susceptibility measurements,<sup>7</sup> the ratio  $H_K/T_K \approx 5.7$  kG/K requires  $A \approx 1.15$  in Eq. (1). With  $S = \frac{3}{2}$  and  $g = 2$  assumed for the magnetic impurity in  $\text{Cu}_{99.5}\text{Au}_{0.5}\text{-Fe}$ , the ratio  $H_K/T_K = 5$  kG/K obtained in Ref. 1 requires that  $A = 1.0$ . This ratio was obtained for a well-annealed specimen, and it was found that strain in the specimens caused an increase in this ratio to as much as 8 kG/K. The preparation of the  $\text{Cu}_{98.25}\text{Au}_{1.75}\text{-Cr}_{0.0035}$  ingot required two violent quenches from the melt, and the increased value of  $A$  for this alloy may not be due to intrinsic properties but instead may be a result of the heat treatment.

The presence of the gold constituent in  $\text{Cu}_{98.25}$

$\text{Au}_{1.75}\text{-Cr}_{0.0035}$  allows a reasonably accurate determination of  $\Delta\rho/c$  to approximately 100 K, whereas in the results of Daybell and Steyert, uncertainties in the separation of the electron-phonon part of the resistivity caused large uncertainties in  $\Delta\rho$  at temperatures down to 10 K.<sup>2</sup> Deviations from Matthiesen's rule have been effectively suppressed in the temperature regime below 100 K in Fig. 1 since the peak in such deviations occurs at higher temperatures where the total "impurity" resistivity (both Au and Cr atoms) and the electron-phonon resistivity are comparable. At the same time, judging by results for  $\text{Cu}_{1-x}\text{Au}_x\text{-Fe}$  alloys,<sup>1,8</sup> a reduction of  $T_K$  by roughly 10% from the value for Cu-Cr is expected for  $\text{Cu}_{98.24}\text{Au}_{1.75}\text{-Cr}$ . In Fig. 1, a slight bulge beginning at roughly 5 K occurs in the  $\Delta\rho/c$  vs  $T$  curves, which may be the start of deviations from Matthiesen's rule which reach a maximum at much higher temperatures. This upward bulge appears to be comparable in magnitude to the error bar at 95 K, and the systematic error in  $\Delta\rho/c$  from this source is therefore expected to be comparable to the error arising from all other sources.

In Fig. 1 the field-independent high-temperature limit for  $\Delta\rho/c$  must be between zero and  $6.3 \times 10^{-6}$   $\Omega$  cm/at. %, since  $\Delta\rho/c$  cannot be less than zero and is not expected to rise above the maximum value allowed by the error bar at 95 K. Using the shape of the zero-field resistivity results by Daybell and Steyert for Cu-Cr,<sup>2</sup> which extend down to 0.04 K, the  $H=0$  curve in Fig. 1 should approach a value of roughly  $18 \times 10^{-6}$   $\Omega$  cm/at. % in the low-temperature limit. Assuming  $T_K$  to occur at the half-height of the resistivity step for  $H=0$  means that for the  $\text{Cu}_{98.25}\text{Au}_{1.75}\text{-Cr}$  Kondo system

$$4 \lesssim T_K \lesssim 20 \text{ K.} \quad (2)$$

For the Cu-Cr system these limits should be adjusted upwards by roughly 10%.

A similar determination of  $T_K$  for the Cu-Fe system yields the value 20 K.<sup>1</sup> Here an approximate value rather than a range can be determined because the linear range in  $c$ , for  $\Delta\rho$ , allows Fe concentrations as large as 0.08.<sup>1</sup> This means a much larger value of  $\Delta\rho$  in comparison to the resistivity components of the matrix, and greater accuracy for  $\Delta\rho/c$ .  $T_K$  for the Cu-Cr system is larger than one fifth of the value for the Cu-Fe system.  $T_K$  for the Cu-Cr system is probably less than one half of the value for the Cu-Fe system because the concentration ranges where  $\Delta\rho$  is linear in  $c$  suggest a ratio of radii for the Kondo state  $(0.0035/0.08)^{1/3} = 0.35$ , and a similar ratio for Kondo temperatures. For the Cu-Cr Kondo system, the electrical resistivity results suggest

$$4 \lesssim T_K \lesssim 10 \text{ K.} \quad (3)$$

In this inequality,  $T_K$  means simply the temperature at the half-height of the Kondo resistivity step, rather than any more basic definitions which might be somewhat different.

Summarizing the results here, and in Ref. 1, the effect of a magnetic field on the Kondo resistivity is similar to the effect of increased temperature. No peak occurs in  $\Delta\rho$  vs  $T$  curves for  $H \neq 0$ .

I have noticed recently that the experimental technique used here and in Ref. 1 to suppress the normal magnetoresistance was suggested for Kondo systems independently and earlier by Abrikosov.<sup>9</sup> I acknowledge the very generous loan of a superconducting magnet and data-acquisition electronics by J. P. Jan, for this work and also for the work in Ref. 1. Specimens were prepared by W. Fisher and G. P. Green, and chemical analysis was performed by members of the Analytic Chemistry Section, National Research Council of Canada.

<sup>1</sup>E. W. Fenton, Phys. Rev. B 5, 3788 (1972); 7, 3144 (1973).

<sup>2</sup>M. D. Daybell and W. A. Steyert, Phys. Rev. Lett. 20, 195 (1968).

<sup>3</sup>R. M. More and H. Suhl, Phys. Rev. Lett. 20, 500 (1968).

<sup>4</sup>P. E. Bloomfield, R. Hecht, and P. R. Sievert, Phys. Rev. B 2, 3714 (1970).

<sup>5</sup>W. B. Pearson, Philos. Mag. 46, 920 (1955).

<sup>6</sup>A. A. Abrikosov, Physics (N.Y.) 2, 5 (1965).

<sup>7</sup>L. Creveling (private communication referred to in Ref. 2).

<sup>8</sup>J. W. Loram, T. E. Whall, and P. J. Ford, Phys. Rev. B 2, 857 (1970).

<sup>9</sup>A. A. Abrikosov, Physics (N.Y.) 2, 61 (1965), the second from the last sentence in this paper.