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LOW-TEMPERATURE RESISTANCE MINIMUM AND MAGNETORESISTANCE FOR DILUTE ALLOYS OF IRON IN COPPER

by John S. Loomis and Wayne R. Hudson

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ABSTRACT

The temperature dependence of the Fe-Cu resistivity was compared with the predictions of Kondo and Nagaoka. The Nagaoka theory fits the experimental results better than the Kondo theory. The transverse magnetoresistance was measured as a function of temperature and magnetic field. For concentrations less than 0.03 at. % Fe in Cu, the magnetoresistance was positive and was approximately linear for higher magnetic fields. For Fe concentrations greater than 0.03 at. %, the magnetoresistance became increasingly negative with increasing concentrations. The magnetoresistance of the 0.073-at.%-Fe sample fit a magnetic field dependency of $H^{1.33}$.

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SUMMARY

The electrical resistance of wire samples of several dilute alloys of iron in copper was measured for low temperatures between 4.2 and 25 K, and for magnetic fields up to 4.5 teslas. The resistance minimum was observed near 20 K. The temperature dependence of the resistivity was compared with the predictions of Kondo and Nagoaka. The Nagaoka theory fits the experimental results better than the Kondo theory. The transverse magnetoresistance was measured as a function of temperature and magnetic field. For concentrations less than 0.03 atomic percent iron (Fe) in copper (Cu), the magnetoresistance was positive and was approximately linear for higher magnetic fields. For iron concentrations greater than 0.03 atomic percent, the magnetoresistance became increasingly negative with increasing concentrations. The magnetoresistance of the 0.073-atomic-percent-Fe sample fits a magnetic field dependency of $H^{1.33}$

INTRODUCTION

In pure metals, the electrical resistance is the result of scattering of electrons by thermal vibrations and crystal imperfections. Above about 50 K, thermal scattering is the dominant factor. As the temperature decreases, the resistance decreases rapidly, typically with a temperature dependence of T^3 or T^5 (ref. 1). At some temperature below 20 K, metals either become superconducting or their resistance approaches a residual value because of impurities and dislocations in the sample. In this temperature range, impurity scattering dominates thermal scattering. Dilute alloys generally behave similarly to pure metals, but in some dilute alloys, the resistance may pass through a minimum and, perhaps at a lower temperature, a maximum. Much is already known about the resistance minimum (refs. 1 and 2). The minimum results from the presence of a solute metal, which is usually a transition metal, such as iron (Fe), manganese (Mn), chromium (Cr), nickel (Ni), etc.

The mechanism that explains the resistance minimum is currently of considerable theoretical interest. Yosida (ref. 3) was the first to try to explain the anomolous resistive behavior as exchange interactions between conduction 4s electrons and the localized 3d electrons. His treatment did not yield a resistance minimum, but it did predict a resistance proportional to the square of the magnetization. Several experiments have since confirmed this result (refs. 4 and 5).

Kondo (ref. 6) extended Yosida's model to a second Born approximation and obtained an s-d interaction resistance with a temperature dependence of $-\ln T$. This term, when added to the lattice resistance, gave a resistance minimum that agreed reasonably well with existing experimental results.

Recently, Nagaoka (ref. 7) found that Kondo's perturbation theory breaks down in the third order. He then studied the s-d exchange interaction using the method of retarded double time Green's functions (ref. 8). Nagaoka found that, for antiferromagnetic interactions, a bound state is formed between the localized-electron spin and the conduction-electron spin. The Nagaoka resistivity has a temperature dependence of the form

$$\left[1 + 2.029 \frac{T^2}{(T_{min} - T)^2}\right]^{-1}$$

where T_{min} is the temperature of the resistance minimum. Daybell and Steyert (ref. 9) reported that the Cu-Fe alloys (0.0022 at. % Fe, 0.0063 at. % Fe, and 0.0560 at. % Fe) that they have tested confirm Nagaoka's resistivity-temperature prediction. In apparent contradiction, Monod (ref. 5) presented data for Cu-Fe alloys, which are fitted by Kondo's $-\ln T$ relation in the temperature range of 4 to 19 K. Monod also found that the negative magnetoresistance of both Cu-Fe and Cu-Mn alloys varies as the magnetic field H to the n^{th} power, where $n = 1.7$ for Cu-Mn and 1.75 for Cu-Fe. He also observed that a low temperature maximum occurred when the magnetic field was applied. The temperature at which the minimum occurred increased as the magnetic field was increased. Muto and Noto (ref. 10) found that the magnetoresistance of the Cu-Fe alloy changed from positive to negative at approximately 0.04 atomic percent Fe in Cu.

Two of the parameters that characterize the resistance minimum are the temperature of the minimum T_{min} and the depth of the minimum $(R_{min} - R_{4.2K})/R_{room}$. For the Cu-Fe system, Pearson (ref. 11) found that T_{min} varies as the 1/5.2 power of the iron concentration, which was in agreement with Kondo's result. Knook (ref. 12) obtained a power dependence of 1/5.3 and found, in addition, that the depth varies linearly in concentration.

The purpose of the research reported herein was to investigate further the electrical resistance of dilute alloys of Fe in Cu as a function of concentration, temperature, and transverse magnetic field. The range of concentrations chosen included the concentrations for which the system passed from positive to negative transverse magnetoresistance (0.01 to 0.10 at. % Fe). The experimental apparatus and techniques were checked by comparing the concentration dependence of the minimum temperature and the depth of the minimum with published results (refs. 11 and 12).

The goals of this investigation were fourfold. The first was to compare the temperature dependence of the resistance with the predictions of Kondo and Nagaoka. The second was to determine the Fe concentration where the sign of the magnetoresistance changed and to compare it with Muto and Noto's result of 0.04 atomic percent Fe. The third was to measure the magnetoresistance for several concentrations and to relate it with Monod's result of $\Delta R \alpha H^{1.75}$ for 0.0110 atomic percent Fe. The fourth was to extend Monod's data on the magnetic field dependence of resistance against temperature in an effort to obtain a low-temperature maximum for Cu-Fe.

SYMBOLS

H	magnetic field, tesla
R	resistance, $\mu\Omega$
T	temperature, K
x	iron concentration, at. %
Subscipts:	
min	minimum
room	room temperature
Superscript:	
n	power dependency

APPARATUS

A variable temperature probe (see fig. 1) was designed to fit into the bore of a 5-tesla superconducting solenoid. Temperature control was established by balancing the current to a resistance heater against a controlled heat leak to the liquid helium out-

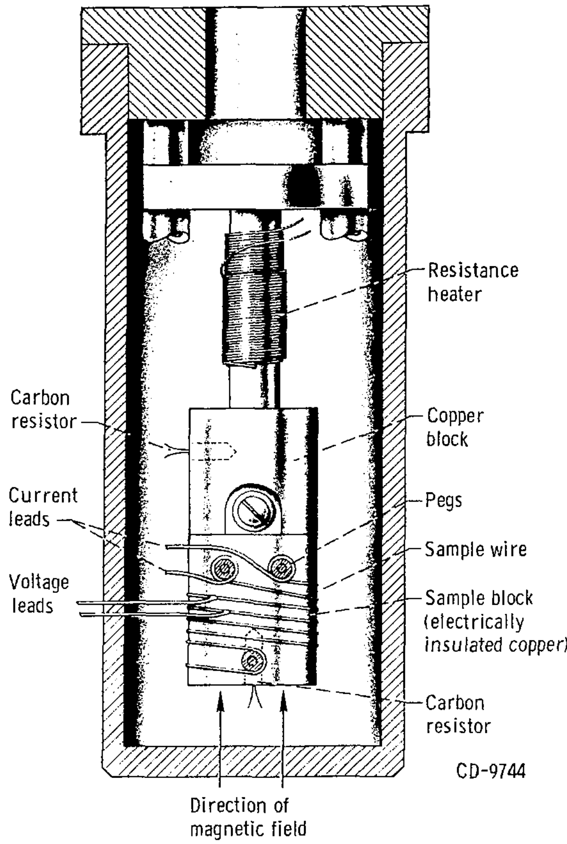

Figure 1. - Design details of experimental probe.

side the probe. The sample was mounted on a detachable portion of the copper base, which had been covered by a thin tape for electrical insulation. The sample itself was coated with varnish to keep it fixed and electrically insulated. Two calibrated, 100 ohms, 0.1 watt, carbon resistors were mounted in the copper block to serve as temperature sensors. Calibration points at the liquid-helium boiling point (4.2 K), the liquid-hydrogen boiling point (20.2 K), and the liquid-nitrogen boiling point (77 K) were fitted to an empirical equation developed by Clement and Quinnell (ref. 13).

Sample voltages were amplified by the amplifier section of a nanovoltmeter and displayed on either an X, Y-recorder or a digital voltmeter. Other measurements, such as sample current, heater current, and thermometer voltage could be displayed directly on the X, Y-recorder or digital voltmeter. Current sources were constant to within ±0.05 percent. Bucking circuits were employed to allow measurements of small differences in the sample voltage. The magnetic field was determined by measurement of the voltage output of either the magnet current shunt or a Hall probe, after which the output was compared with calibrations of the shunt and the Hall probe.

The alloys were prepared commercially and drawn into the sample wires. The source of the 0.0105-atomic-percent-Fe alloy is unknown. The other alloys were prepared from three-pass electron-beam zone-refined iron and vacuum-melted copper alloyed in an arc melter. All sample wire was 0.051 centimeter (0.020 in.) in diameter, and concentrations were measured by a commercial laboratory. The correctness of these measurements relative to each other was verified by measurement of the resistivity at 4.2 K, which depends linearly on the concentration. Separate current and voltage leads were attached to the sample. The test sample preparation consisted of silver soldering voltage taps of the same wire to the sample. This procedure eliminates contact potentials that occur between dissimilar materials. The sample length between the voltage taps was approximately 25 centimeters. Then the sample was cleaned in dilute nitric acid and rinsed. A few samples were annealed after being mounted on the sample block, but no differences in results were observed. All samples were wound bifilarly on the sample block, as illustrated in figure 1.

EXPERIMENTAL PROCEDURE

The average resistance of the sample at room temperature and at 4.2 K was determined by a number of voltage-current measurements. The measured resistances were divided by the resistance at room temperature to obtain a dimensionless resistivity parameter. The standard procedure for continuously monitoring the sample voltage on the X, Y-recorder was to reverse periodically the direction of the sample current (and simultaneously the polarity of the voltage output circuit). This procedure produced two broken curves, one for each current direction. A smooth curve could then be drawn through these broken segments; hence, many of the smaller voltage transients (e.g., when the heater current was increased) could be eliminated. Furthermore, averaging the two curves permitted thermal or stray voltages independent of the current to cancel. Estimates of the reproducibility were obtained by a comparison made of several curves from different sample currents and for different days. The curves of resistance against temperature were reproducible to within 0.5 percent of the resistance at 4.2 K and zero applied magnetic field. The reproducibility of the magnetoresistance measurements was 0.37 percent.

Curves of resistance against temperature were traced out slowly from 4.2 to 25 K in the course of perhaps $1\frac{1}{2}$ to 2 hours to approximate equilibrium conditions as nearly as possible. The curves of resistance as a function of transverse magnetic field were obtained similarly. Here the temperature was monitored continuously by the voltage drop across a carbon resistor and was displayed on the digital voltmeter. Magnetoresistance effects in the carbon resistor were noted, but because they were less than 3 percent, no temperature corrections were made.

RESULTS AND DISCUSSION

Some of the measured properties are summarized in table I. Nordheim's rule for the residual resistivity of a binary alloy containing a mole fraction x of one element and (1 - x) of a second element is $\rho_r(x) \propto x(1 - x)$ (ref. 1, p. 337). For low concentrations, this equation yields a linear relation. The good agreement between Nordheim's rule and the present experimental results is shown in figure 2; this agreement lends credence to the accuracy of the concentrations presented in table I.

TABLE I. - SAMPLE PARAMETERS, DEPTH, AND TEMPERATURE OF RESISTANCE MINIMUM

Nominal iron concentration, x, at. %	Resistance at room temperature, R_{room}, mΩ	Resistance at 4.2 K, $R_{4.2\,K}$, mΩ	Resistance ratio at 4.2 K, $R_{room}/R_{4.2}$	Temperature at minimum resistance, T_{min}, K	Depth of resistance minimum, $(R_{min} - R_{4.2\,K})/R_{room}$, dimensionless
0.0105	27.10	2.267	11.9	14.8	-7.1×10^{-3}
.018	24.55±0.013	3.29	7.46	17.0	-11.7
.032	25.04±0.01	4.55±0.01	5.50	18.0	-17.2
.045	25.68±0.03	5.83±0.03	4.40	19.5	-23.4
.073	30.25±0.04	10.89±0.01	2.78	23.1	-36.5

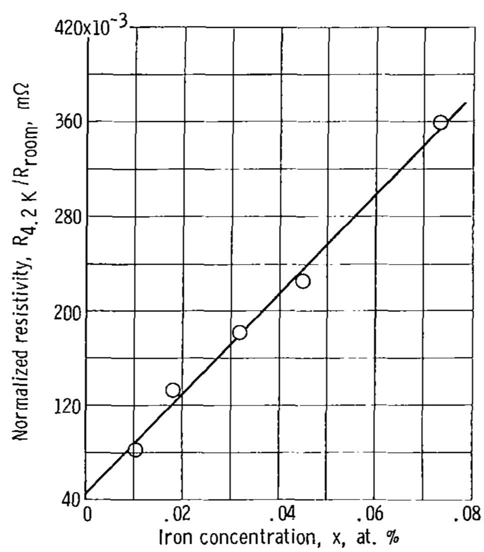

Figure 2. - Resistivity as function of iron concentration at 4.2 K.

Figure 3 shows the change in resistivity as a function of temperature from 4.2 to 34 K for the different alloys. The experimental apparatus and techniques were checked

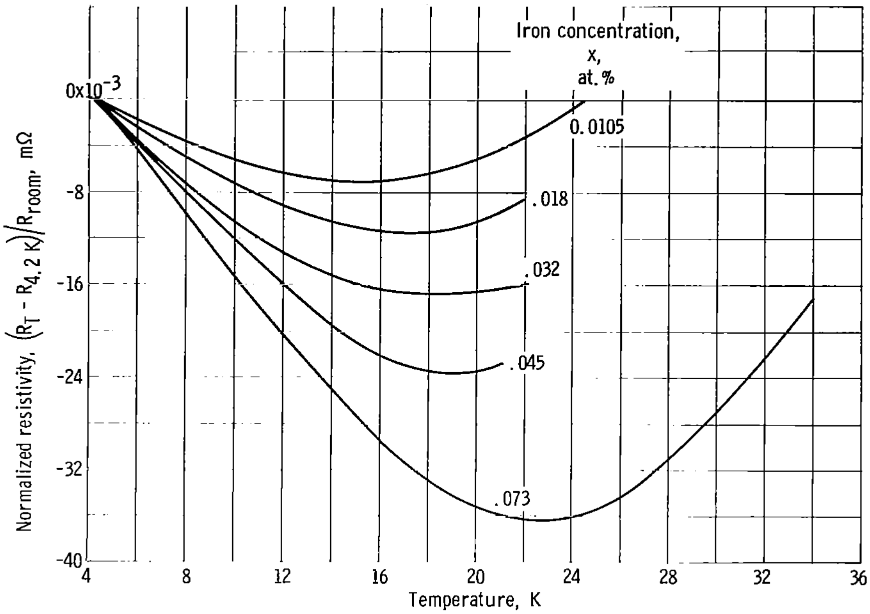

Figure 3. - Resistivity as function of temperature for several iron concentrations.

by comparing the concentration dependence of the minimum temperature and the depth of the resistance minimum

$$\frac{R_{min} - R_{4.2\,K}}{R_{room}} \times 10^3$$

with published results (refs. 11 and 12). If the minimum temperature T_{min} is plotted against the concentration x on logrithmic scales, as in figure 4, a straight line is obtained, the slope of which was determined by a least-squares fit. The calculated slope and the measured values of x and T_{min} were used to determine the average value of the following equation relating x and T_{min}

$$T_{min} = 40.1\, x^{1/4.53}$$

where

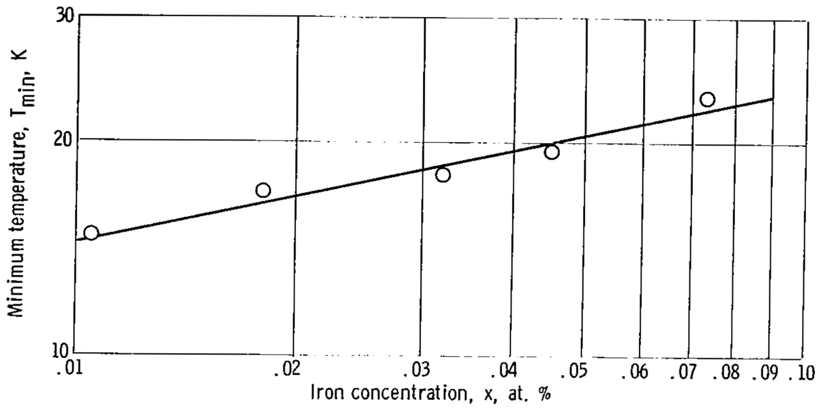

Figure 4. - Minimum temperature as function of concentration.

where T_{min} is in kelvin and x is in atomic percent of Fe. This result compares within the experimental accuracy with Knook's result (ref. 11), which is

$$T_{min} = 43.7\ x^{1/5.3}$$

The depth of the minimum increases linearly, as shown in figure 5. This increase is also in agreement with that shown by Knook (ref. 12).

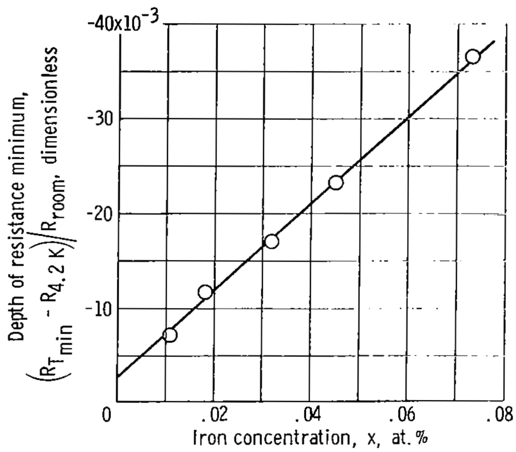

Figure 5. - Depth of resistance minimum as function of iron concentration.

The experimentally obtained curves of resistivity against temperature (fig. 3) are compared with the predictions of Kondo and Nagaoka in figures 6 and 7, respectively. The Kondo theory (fig. 6) fits the data best for the lower iron concentrations and the higher temperature region. The Nagaoka theory (fig. 7) fits the data well at low temper-

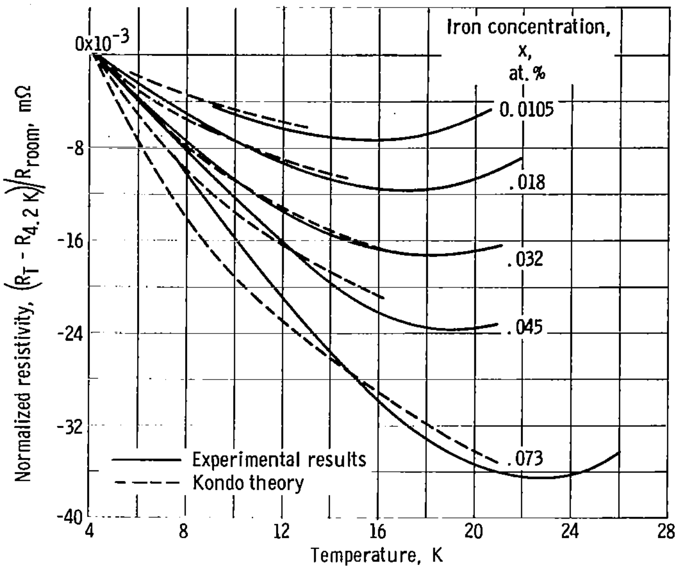

Figure 6. - Comparison of experimental results and Kondo theory (ref. 6) for resistivity as function of temperature for several iron concentrations.

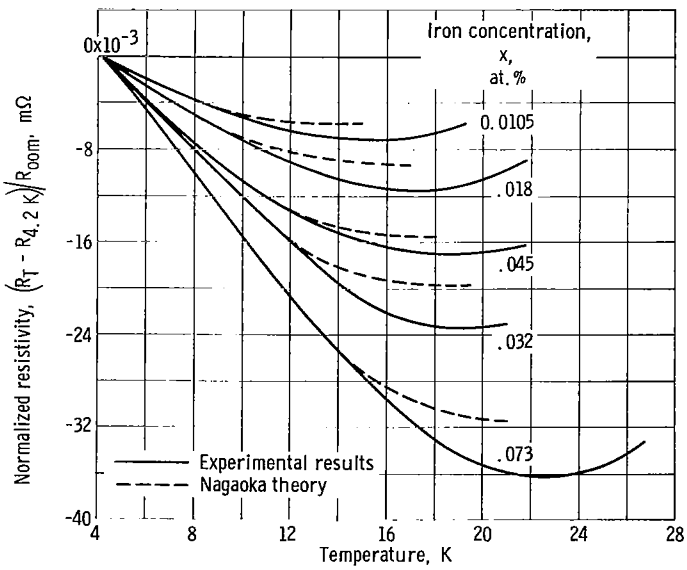

Figure 7. - Comparison of experimental results and Nagaoka theory (ref. 7) for resistivity as function of temperature for several iron concentrations.

atures for all five alloys. In the upper portion of the temperature range, the Nagaoka theory does not predict as large a decrease in resistance as observed. Over the whole range of concentrations and temperature, the Nagaoka theory fits the results better than the Kondo theory.

The third experimental purpose was to determine the Fe concentration at which the magnetoresistance changes sign. Figure 8 gives the magnetoresistance data for the five

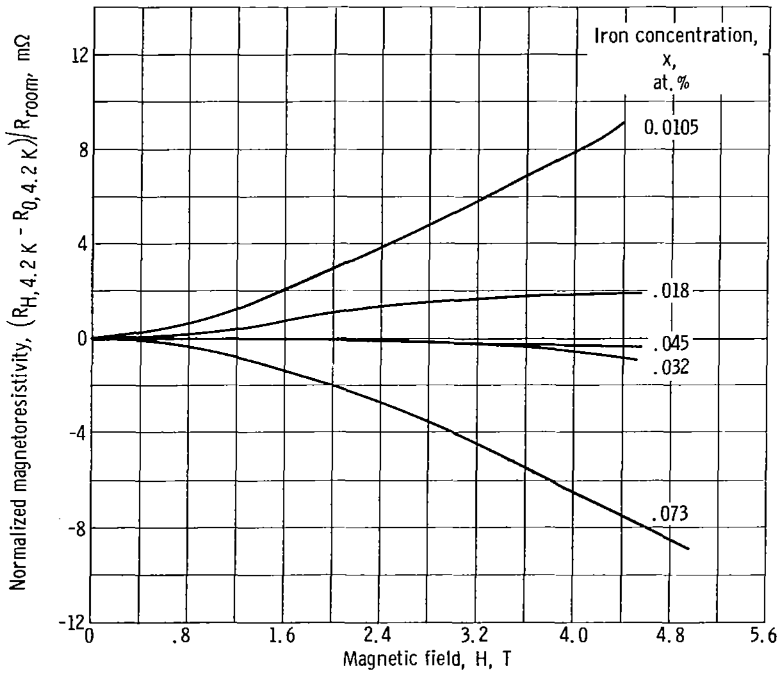

Figure 8. - Magnetoresistivity as function of transverse magnetic field at 4.2 K for several iron concentrations.

alloys. The concentration at which the magnetoresistance changed sign appeared to be between 0.032 and 0.018 atomic percent Fe. This range may be compared with the range between 0.049 and 0.021 atomic percent Fe obtained by Muto and Noto.

For the 0.073-atomic-percent-Fe alloy at 4.2 K, the logarithm of the magnetoresistance was plotted against the logarithm of the magnetic field. The data were fitted almost perfectly by $\Delta R \propto H^{1.33}$. Monod (ref. 5) obtained a result of $\Delta R \propto H^{1.75}$ for a 0.0110-atomic-percent-Fe sample. The difference between the two measurements probably results from internal magnetic fields that were present in the 0.073-atomic-precent-Fe sample because of the higher iron concentration.

The curves for the magnetic field dependency of the resistance against temperature are shown in figure 9. The effect of the magnetic field is to decrease the resistance. The resistances converge as the temperature increases, until at about 25 K where the curves become one, within experimental error. No sign of a maximum in the resistance in the low-temperature region was observed at 4.2 K and 4.0 teslas. The reason may be that the Cu-Fe system only exhibits the maximum phenomena at much higher magnetic fields, if at all.

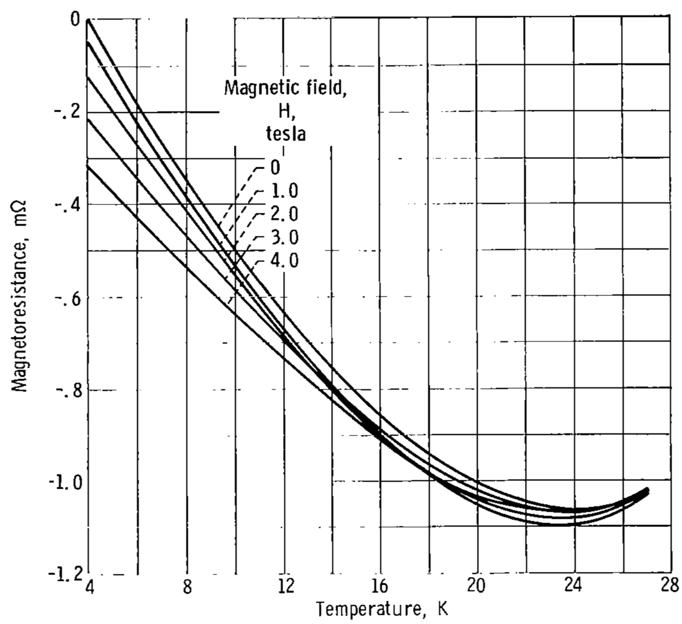

Figure 9. - Magnetoresistance as function of temperature for various magnetic fields. Iron concentration, 0.073 at.% Fe.

CONCLUDING REMARKS

At low temperatures, the experimentally determined temperature dependency of resistivity agreed well with the predictions of Nagaoka. The concentration at which the magnetoresistance changed sign was determined to be between 0.032 and 0.018 atomic percent Fe. The magnetoresistance of the 0.073-atomic-percent-Fe sample was proportional to $H^{1.33}$. Monod's (ref. 5) result for a 0.0110-atomic-percent-Fe sample was $H^{1.75}$. The magnetic field dependency for the curves of the resistance against the temperature was extended to 4.0 teslas. No indication of a low-temperature maximum was observed.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, April 2, 1968,
129-02-05-14-22.

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