The Hall Effect of Copper-Nickel Alloys*

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(Received September 11, 1952)

The ordinary and extraordinary Hall effects have been measured at room temperatures for the Ni—Cu series of alloys. The number of conduction electrons calculated from $n^*=1/R_0Nec$ (where R_0 is the "ordinary" constant measured at high fields) agrees fairly well with the number of 4*s* electrons predicted for the alloys from the simple band model. It is shown that the agreement for these alloys should improve at lower temperatures. Results in pure nickel and copper do not agree so well. A two-band model is invoked to explain the ordinary effect in pure nickel. Since in pure copper the 4*s* band is half-full, the simple approximation, valid for nearly empty bands, should not be expected to hold. No explanation is offered for the extraordinary Hall effect in any of the samples.

A. INTRODUCTION

IT has been shown,[1–4] that the Hall emf in ferromagnetics can be separated into two distinct parts, one proportional to H (the ordinary Hall effect) and one proportional to M (the extraordinary Hall effect). It has been proposed[3] that e_H, the ferromagnetic Hall electric field per unit current density be written as

$$e_H=E_H/ib=R_0(H+\alpha 4\pi M), \qquad (1)$$

where E_H = the Hall emf, R_0 = the ordinary Hall constant, i = the current density in the ferromagnetic sample, b = the width of the sample between the Hall probes, M = the intensity of magnetization, and H = the magnetic field corrected for demagnetization. Values of R_0 and α have been determined previously for nickel.[3] This paper reports experimentally determined values of R_0 and α for the nickel-copper alloy system at room temperatures.

The nickel-copper system was chosen for several reasons. First, copper alloyed with nickel forms a homogeneous single-phase substitutional solid-solution over the entire range of composition. Second, the nickel-copper system saturates at low magnetic fields, and only those materials having saturation magnetizations smaller than the field of the magnet used could be investigated. Third, the number of holes in the 3*d* band of this alloy system and, consequently, the number of electrons in the 4*s* band have been quite accurately determined from measurements of the saturation magnetization.[5,6] One expects the ordinary Hall constant to be a measure of the number of electrons taking part in conduction processes. According to the simple band model, the addition of copper to nickel should increase the number of electrons in the 3*d* band until this band is filled, and, thereafter, should increase the number of electrons in the 4*s* band.

The Hall constant may be defined by $R=\partial e_H/\partial B$, where R is independent of B in most materials but not in the ferromagnetic materials. The R_0 which is independent of B in ferromagnetic materials can be obtained from the slope of the e_H *vs* B curve at high fields where $M=M_s=$ constant. From Eq. (1), since $H=B-4\pi M$,

$$\partial e_H/\partial B=R_0\{1+4\pi(\alpha-1)\partial M/\partial B\}, \qquad (2)$$

which gives $R_0=\partial e_H/\partial B$ when $\partial M/\partial B=0$. If B_s is the smallest value for B for which $M=M_s$, then above B_s the e_H *vs* B curve is a straight line. Extrapolating this straight line back to $B=0$ gives $4\pi(\alpha-1)R_0M_s$, from which α can be obtained provided M_s is known. Thus, R_0 is obtained from measurements of the change of E_H with incremental changes in field above saturation and α is obtained from measurements of the change in E_H on reversal of any given field above $B=B_s$.

B. METHOD OF MEASUREMENT

Measurements were made on nickel-copper alloys containing 0, 10, 20, 30, 40, 50, 60, 80, 90, 95, and 100 percent copper. Each sample was measured in the unannealed state and after it had been annealed for two hours. The samples were 4.5 cm×2.0 cm×0.1 cm and were mounted in the holder shown in Fig. 1. They were hard soldered into two copper electrodes, which were fastened to micarta strips for added rigidity. The Hall

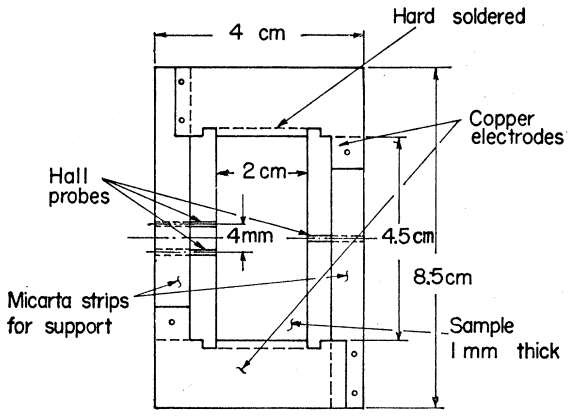

FIG. 1. Sample holder for Hall emf measurements.

* This research was supported by the ONR.
† Now at the Naval Research Laboratory, Washington, D. C.
[1] A. W. Smith and R. S. Sears, Phys. Rev. **34**, 1466 (1929).
[2] Emerson M. Pugh, Phys. Rev. **36**, 1503 (1930).
[3] Pugh, Rostoker, and Schindler, Phys. Rev. **80**, 688 (1950).
[4] N. Rostoker and E. M. Pugh, Phys. Rev. **82**, 125 (1951).
[5] C. Sadron, Ann. Physik **17**, 371 (1932).
[6] H. Krutter, Phys. Rev. **48**, 664 (1935).

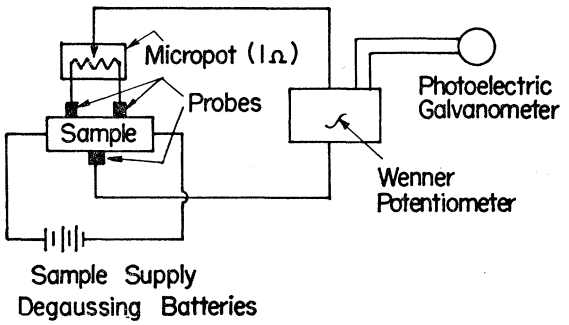

FIG. 2. Circuit diagram for Hall emf measurements.

probe connections shown in Fig. 2 were adapted from a method originated by Kolacek.[7] One probe on one side and two probes (4 mm apart) on the other side of the samples were used. The two probes on the same side were connected to a 1-ohm Micropot, used as a voltage divider. The center tap of this Micropot and the remaining Hall probe were then connected to a Wenner potentiometer with a Rubicon photoelectric galvanometer which has a linear response for small deflections. The Wenner potentiometer is graduated into steps of 10^{-7} volt, but interpolations can be made with the galvanometer deflections to $\pm 5\times 10^{-9}$ volt.

Copper-constantan thermocouples were connected to each Hall probe to determine the temperature of the sample and to determine the corrections for the Ettingshausen effect. The field was measured with a flat spiral shaped flux coil in contact with the sample and connected to a greatly overdamped high sensitivity galvanometer operating as a fluxmeter. The sample with flux coil and thermocouples was placed between the poles of the electromagnet, with the flat surface perpendicular to the field. Glass wool wrapped around the whole assembly prevented air currents from disturbing the thermal equilibrium. To change the magnetic field without disturbing the temperature equilibrium, a method previously devised by one of us[8] was adapted for use with the available Kohl magnet. By reversing the direction without changing the magnitude of the current in a selected coil the field was changed without affecting the Joule heating. This greatly improved the stability of the readings. Since the four coils of the Kohl magnet had approximately equal numbers of turns it was necessary to shunt three coils with different resistors to insure that different field changes would be produced by reversing different coils. The resistors were chosen to provide five convenient steps in magnetic field above the saturation of the samples for the R_0 determinations. To avoid breaking the current during these reversals, each coil reversing switch was made to produce a temporary short instead of an open circuit across the coil at the mid-point in its reversal. Since the four coils were in series, one coil could be temporarily shorted without causing difficulty.

To insure sufficient stability for the required accuracy good thermal equilibrium was essential. The magnet and sample currents were turned on for three hours before measurements were to be made. High stability during measurements was also required for these currents. For the magnet current, a 120-volt storage battery was floated across a 220-volt dc line. Four Navy degaussing type batteries in parallel supplied the large sample current that was required and maintained it quite steady for several hours.

To make certain that the true Hall effect is measured, one must eliminate all thermal emf's, especially those due to the Ettingshausen effect. Most of the thermal emf's were eliminated by maintaining temperature equilibrium throughout the measuring circuit and then observing the changes produced by changes in the magnetic field. However, the Ettingshausen effect causes changes in the temperatures at the Hall probes with changes in the magnetic field. The thermocouples connected to the Hall probes were used to determine these field dependent temperature changes. In all cases the corrections were small.

C. RESULTS AND DISCUSSION

The curves in Fig. 3(a) and 3(b) show the variation in Hall voltage with the magnetic induction (beyond

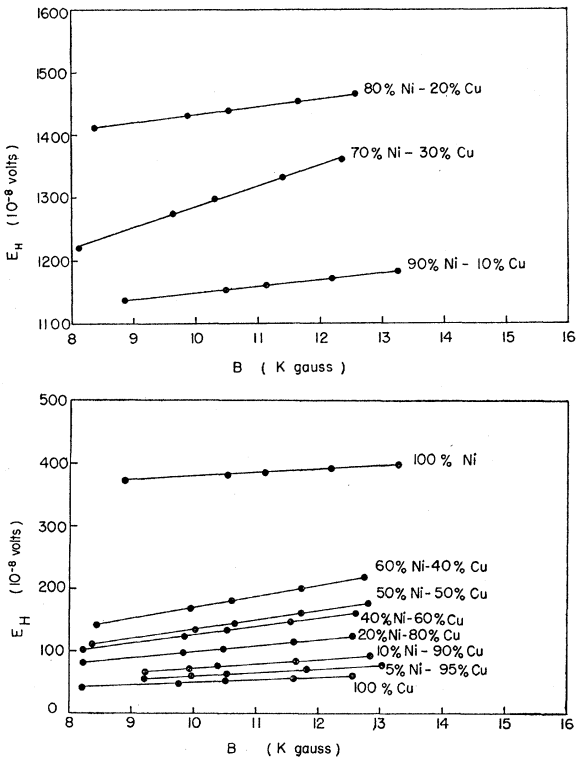

FIG. 3. (a) Hall emf *versus* magnetic induction, using 10 amp through 0.1-cm thick samples of Ni—Cu alloys at 20°C. (b) Hall emf *versus* magnetic induction, using 10 amp through 0.1-cm thick samples of Ni—Cu alloys at 20°C.

[7] F. Kolacek, Ann. Physik **39**, 1491 (1912).
[8] See reference 2, p. 1506.

saturation) for the different alloys of copper and nickel using a 10-amp sample current at 20°C. Each point on the curves represents an arithmetical average of five runs. Figure 4 illustrates the variation of R_0 with copper content, while Fig. 5 shows the variation of n^* ($n^*=1/NR_0ec$) with copper content. Values for the field parameter α at 25°C are plotted *versus* copper content in Fig. 6. The spread of the data is such that the standard deviations in the R_0's are in the neighborhood of 2 percent, except for the 30 percent Cu alloy. For this alloy the spread is greater because its Curie point is very close to the temperature of the measurements.

The values of n^*, as shown in Fig. 5, are close to the number, n_s, of electrons in the 4*s* conduction band predicted from magnetic measurements[5,6] using the simple band model. The predicted values of n_s are shown in Fig. 5 by the dotted line. According to these predictions, adding copper to nickel adds electrons to the 3*d* band and not to the 4*s* band until the 3*d* band is filled at 60 percent copper. From this point on the extra electrons fill up the 4*s* band.

When values of R were determined from measurements at low fields as they have been in the past, the values of $n=1/NRec$ differed in order of magnitude from predicted values of n_s, for most ferromagnetic materials. Here, with R_0 obtained from high field measurements, the calculated values of n^* do not differ from the predicted values of n_s by as much as a factor of 2. In some of the alloys the agreement is quite striking. The agreement is even more striking when it is realized that the measurements on the alloys containing 20 and 30 percent copper were measured at room temperatures which are close to their Curie points where $\partial M/\partial B\neq 0$ at the highest fields available. If a correction[9] is made for this, the experimental values for these

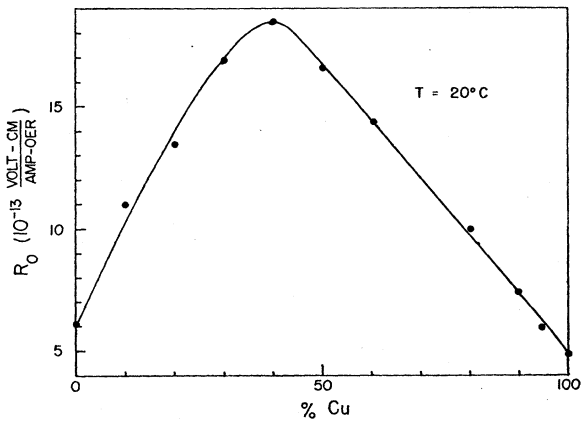

FIG. 4. Ordinary Hall constant, R_0, *versus* Cu content in Ni—Cu alloys at 20°C. Not corrected for the fact that $\partial M/\partial B\neq 0$ above technical saturation.

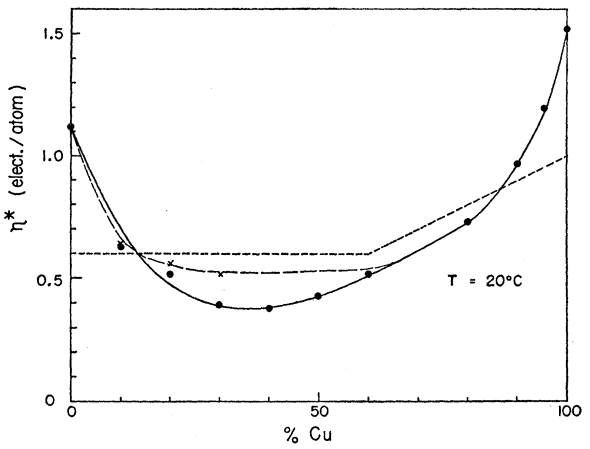

FIG. 5. Values of $n^*(1/NR_0ec)$ *versus* Cu content in Ni—Cu alloys at 20°C. Values taken directly from the slopes of the lines in Fig. 3 are shown as circles connected with a solid line. Values obtained by correcting for the fact that $\partial M/\partial B\neq 0$, even at high fields, are shown as crosses connected by a line of long dashes; which is extended smoothly through the 40 percent and the 50 percent Cu alloys, since corrections of similar magnitude but of unknown value should be made for them also. The numbers of 4*s* electrons per atom, predicted by simple band theory, are indicated by the line of short dashes.

two alloys move up toward the theoretical curve. Unfortunately, the values of $\partial M/\partial B$ are not known for these alloys at the temperatures of these measurements. It is known, however, that the correction due to this factor is of the right magnitude[9] to improve this agreement for these alloys.

The study made by Rostoker[4] and one of us of the Hall effect measurements on Ni made by Smith[10] at temperatures above and below its Curie point, suggests that similar corrections should be applied for the alloys having 40 and 50 percent Cu. A correction here would also improve the agreement between theory and experiment for these alloys. With the 60, 80, and 90 percent copper alloys the agreement between theory and experiment is fairly good. Low temperature measurements now being made on this series of alloys may clear up these discrepancies.

The field parameter α can be calculated from the data for those samples that are ferromagnetic at room

[9] Since the completion of the experimental part of this paper, the order of magnitude of this correction has been determined by Norman Rostoker and Simon Foner by assuming that $M/M_s=f(T/T_c, \mu H/kT)$ has the same form for all ferromagnetics.

The saturation magnetizations, M_s, and the Curie temperatures, T_c, are known for all of the alloys and for pure nickel. Since accurate measurements of M in nickel have been made by P. Weiss and R. Forrer [Ann. phys. 5153 (1926)] for a wide range of values of T and H, empirical values for $f(T/T_c, \mu H/kT)$ could be obtained from the nickel data and applied to the alloys. Values of M *versus* H (therefore, M *versus* B) for the alloys at the temperature of the measurements were estimated from the nickel data. From these, values of $\partial M/\partial B$ were obtained and used in Eq. (2) to obtain corrected values of R_0 from the high field data. The corrected values of n^* are shown in Fig. 5 by crosses connected with a dashed line. The nickel measurements of M do not extend to sufficiently high temperatures to make it possible to use this method for obtaining the corrections for the 40 percent and 50 percent copper alloys, whose Curie temperatures are below the temperature of these measurements. However, the smooth behavior of R_0 and α in nickel (see reference 4) when passing through the Curie point appears to justify drawing the dashed line smoothly to the nonmagnetic alloy (60 percent Cu, 40 percent Ni).

[10] A. W. Smith, Phys. Rev. **30**, 1 (1910).

TABLE I. Values of R_0 and α for those alloys that are ferromagnetic at 20°C. The right-hand columns of both R_0 and α are corrected for the fact that $\partial M/\partial B \neq 0$ even at high fields.

Alloy comp % Cu	$R_0\alpha\left(10^{-13}\frac{\text{volt-cm}}{\text{amp-oer}}\right)$	$R_0\left(10^{-13}\frac{\text{volt-cm}}{\text{amp-oer}}\right)$		α	
		Uncorr.	Corr.	Uncorr.	Corr.
0	60.7	6.07	6.07	10.0	10
10	235	11.0	10.8	21.4	22
20	396	13.4	12.3	29.6	32
30	795	16.9	12.9	47	62

temperatures; namely, the alloys containing 0 percent, 10 percent, 20 percent, and 30 percent Cu. The product $R_0\alpha$ for each alloy is obtained from the intercept of its straight line with the $B=0$ axis in Fig. 3 and its saturation magnetization. These values are shown in Table I. The value calculated for α then depends upon the value of R_0 chosen. Table I shows two values of R_0; the first has been obtained directly from the slopes of the lines in Fig. 3 and the second has been corrected[9] for the fact $\partial M/\partial B \neq 0$ even at high fields. Table I also show values of α obtained both with the corrected and the uncorrected values of R_0.

Since the n^* curve crosses the predicted curve of n_s fairly close to the alloy concentration at which this series changes from being paramagnetic to being diamagnetic, one is tempted to speculate on the possibility that a correction of the opposite sign due to a change in the sign of $\partial M/\partial B$ would improve the agreement for pure copper. However, for such a correction to account for the fact that in copper $n^*=1.48$ would require an α of the order of 10^5. This does not seem likely.[11] A more plausible explanation is that, since in copper the $4s$ band is half-full, the simple approximation of the spherical Fermi surface implicit in $R_0=1/Nnec$, which is valid for nearly empty bands, does not hold.

Two possible explanations have been proposed[3] for the large value of n^* (or small value of R_0) in pure nickel. One of these assumes that R_0 is reduced by conduction in the $3d$ band. This $3d$ conduction would tend to produce a positive R_0 while the $4s$ conduction produces a negative R_0. This explanation seems plausible, since $R_0\alpha$ is known, from experiments of Smith[10] and others, to be positive for both cobalt and iron. $R_0\alpha$ passes through zero at some alloy of cobalt and nickel. If this point of view is adopted, then

$$R_0=-(\sigma_s^2/n_s\sigma^2-\sigma_d^2/n_d\sigma^2)/Nec,$$

and the ratios of the conductivities due to the $3d$ and $4s$ bands can be determined. For pure nickel $\sigma_d/\sigma_s=0.3$ and, for Ni 90 percent and Cu 10 percent, $\sigma_d/\sigma_s=0.03$. This appears to be the most logical explanation for the large value of n^* in nickel.

No satisfactory explanation for the value of α is available. A paper by Rostoker[12] and one of us reviews the various attempts to explain the values for α, which are strongly dependent on temperature, and points out the inadequacies of each of them.

The authors are indebted to the ONR for funds to carry on this research. They are also indebted to J. Goldman, R. Smoluchowski, N. Rostoker, and Simon Foner for assistance and stimulating discussions. They wish to thank the International Nickel Company for furnishing the nickel-rich alloys and the Westinghouse Manufacturing Company for furnishing the copper-rich alloys.

[11] It is interesting to notice that n^* from Hall measurements is generally less than expected in paramagnetic elements (e.g., Li, Na, Mg) and greater than expected in diamagnetic elements (e.g., Cu, Ag, Au). The values of α required to account for these discrepancies are very large.

[12] Presented to the Conference on Magnetism at the University of Maryland, September 2 to 6, 1952. It is planned that the papers from this conference will appear in the *Reviews of Modern Physics*.