

equation for the energy of the surface states for the crystal delimited by the straight line $m_2=0$:

$$g_1(x) = \frac{1}{\gamma'^2 x} (x^2 - 2\gamma'x - 1)(x^3 - 3\gamma'x^2 - x + \gamma') = |b|^2, \quad (55)$$

where the quantity b is defined by Eq. (39).

The condition for the existence of surface states then reads

$$g_2(x) < g_1(x) < 6, \quad (56)$$

if the function $g_2(x)$ is defined as follows:

$$g_2(x) = \frac{1}{\gamma'^2} [x^3 - 3\gamma'x^2 - x + \gamma']^2. \quad (57)$$

With regard to the similarity of the results obtained for the σ bonds of the two-dimensional graphite model with the results for diamond, we give here no detailed discussion of the spectra of the surface states produced by breaking of the σ bonds in graphite. For orientation it will be probably sufficient to refer to Fig. 5 for $\gamma'=0.25$.

Hall Effect and Resistivity of Ni-Pd Alloys*†

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The two Hall coefficients and the resistivity of Ni-Pd alloys have been measured from 4°K to room temperature using fields up to 3.1 webers/m². The ordinary Hall coefficient is found to decrease in magnitude for small additions of Pd in Ni, but to increase as more Pd is added. It is also found that the ordinary Hall coefficient varies more slowly with composition for these alloys than it does for the Ni-Cu alloys. It is shown that a simple treatment successfully correlates the ordinary Hall coefficient, the resistivity, and the saturation magnetization of these alloys. The results indicate that the parallel half of the d band in pure Ni is not quite full at the absolute zero of temperature, and that the relaxation time is not a function only of the energy for the thermal scattering in these alloys. The extraordinary Hall coefficient is found to obey none of the variously proposed theoretical relations.

INTRODUCTION

IT has been established by a number of experiments that the Hall effect in ferromagnetics obeys a relation of the form

$$Vt/I = R_0(\mu_0 H) + R_1 M, \quad (1)$$

where V is the Hall potential, t the sample thickness, I the sample current, H the magnetic field and M the magnetization. R_0 and R_1 have been called¹ the ordinary and extraordinary Hall coefficients, respectively. The ordinary coefficient R_0 was found to correspond to the Hall coefficient in nonferromagnetic materials.

An analysis of the Ni-Cu² and Ni-Co³ data based upon the free electron relation

$$R_0 = -1/(Nne), \quad (2)$$

where N is the number of atoms per m³, n the number of conduction electrons per atom, and e the magnitude

of the electronic charge in coulombs, gave for n_s , the number of $4s$ electrons per atom, values agreeing only within a factor of two with those deduced from magnetic data. Nor could this discrepancy be explained on the basis of a two band model where conduction occurs in both the $3d$ and $4s$ bands.

A four band model, proposed by Pugh,⁴ has been more successful in explaining the data. In this model, both the $3d$ and $4s$ bands are divided into sub-bands where the spins of the electrons are aligned either parallel or antiparallel to the magnetic field. Applications of this model have usually neglected the conduction in the d band, in which case the expression for the ordinary Hall coefficient reduces to

$$R_0 = (-2/Nn_s e) [1 - 2\lambda/(1+\lambda)^2], \quad (3)$$

where λ is the ratio σ_a/σ_p ; σ is the conductivity and the subscripts a and p refer to the antiparallel and parallel electrons, respectively. In terms of the effective number of conduction electrons per atom, n^* , defined by

$$R_0 = -1/(Nn^* e) \quad (4)$$

this reduces to

$$1/n^* = (2/n_s) [1 - 2\lambda/(1+\lambda)^2]. \quad (5)$$

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† Submitted by one of the authors (JAD) in partial fulfillment of the requirements for the degree of Doctor of Philosophy at Carnegie Institute of Technology.

¹ E. M. Pugh, N. Rostoker, and A. Schindler, Phys. Rev. **80**, 688 (1950).

² A. I. Schindler and E. M. Pugh, Phys. Rev. **89**, 295 (1953).

³ S. Foner and E. M. Pugh, Phys. Rev. **91**, 20 (1953).

⁴ E. M. Pugh, Phys. Rev. **97**, 647 (1955).

Allison and Pugh⁵ found that Eq. (5) represented quite well the temperature dependence of three Ni-Cu alloys from 4°K to temperatures above their Curie points. This equation has also been used successfully by Beitel and Pugh⁶ in their discussion of the Hall effect in Fe-Co alloys.

For any reasonable assumptions regarding the conductivities in the two halves of the *s* band, the ratio $\lambda = \sigma_a / \sigma_p$ is limited to the values $0 \leq \lambda \leq 1$, the special case $\lambda = 1$ occurring when the material is no longer ferromagnetic. Thus, Eq. (5) is applicable only to the region $n_s/2 \leq n^* \leq n_s$. If n^* lies outside this range, then it is necessary to include also the contribution due to the *d*-band electrons.

Pugh⁴ and Coles⁷ have pointed out that if R_0 were dependent upon only the electronic structure, then it should show a smooth transition in going from Co through Ni to Cu. But the data⁴ show a distinct cusp at Ni. A similar but smaller cusp is seen also at Co^{3,6} in the series Fe-Co-Ni and at Ag⁸ in its alloys with Pd, Cd, Sn, and Sb. As Coles points out, these results are strong evidence in favor of the suggestion that the Hall effect depends upon the nature of the scattering mechanism, for this is purely thermal in the case of the pure metals while it is only partly so in the case of the alloys. He suggests that the most likely fault is in the assumption that the time of relaxation depends only on the energy.

Thus for the transition elements it is not clear that when R_0 deviates from the expression given by Eq. (3), as it does, for example, in dilute alloys of Cu in Ni, whether this deviation represents only a contribution from *d*-band electrons or whether it may also be due to other factors such as a nonspherical relaxation time. With these thoughts in mind, it has seemed desirable to measure the Hall effect in an alloy system for which there is some chance of separating the two effects, without at the same time changing the electronic structure in a wholly unknown way. Such an alloy system is the Ni-Pd system. We have measured for this system the ordinary and extraordinary Hall coefficients and the resistivity.

THEORETICAL CONSIDERATIONS

The usual one-electron approximation is employed, and Boltzmann's equation is solved subject to the following assumptions:

(i) The conduction electrons can be described by two overlapping bands that, in the Fe, Co, Ni transition elements, arise from the $3d$ and $4s$ atomic states.

(ii) The effect of collisions can be taken into account by a relaxation time that is a function of the energy only.

(iii) The energy of the *s*-band electrons can be represented by

$$E_s = \alpha_s k^m$$

and that of the *d*-band electrons by

$$E_d = A - \alpha_d k^n.$$

Neither m nor n is necessarily 2.

Under these assumptions the solution for the ordinary Hall coefficient is given by⁹

$$R_0 = -\frac{1}{NeL} \left[\frac{1}{n_s} \left(\frac{\sigma_s}{\sigma} \right)^2 - \frac{1}{n_d} \left(\frac{\sigma_d}{\sigma} \right)^2 \right], \quad (6)$$

where n_s and n_d are the number of $4s$ electrons and $3d$ holes, respectively. The electrical conductivities associated with the two bands are designated by σ_s and σ_d , and their sum by σ . As pointed out by Pugh,⁴ however, this analysis does not include any effects arising from the ferromagnetic properties of the Fe, Co, Ni transition elements. With this in mind, he generalized Eq. (6) as

$$R_0 = -\frac{1}{NeL} \left[\frac{1}{n_{sp}} \left(\frac{\sigma_{sp}}{\sigma} \right)^2 + \frac{1}{n_{sa}} \left(\frac{\sigma_{sa}}{\sigma} \right)^2 - \frac{1}{n_{dp}} \left(\frac{\sigma_{dp}}{\sigma} \right)^2 - \frac{1}{n_{da}} \left(\frac{\sigma_{da}}{\sigma} \right)^2 \right]. \quad (7)$$

The additional subscripts p and a divide the *s* and *d* bands each into two sub-bands, where the magnetic moments of the electrons are aligned parallel or antiparallel, respectively, to the magnetic field. It is only above the Curie points of these elements that the two sub-bands are equivalent, in which case Eq. (7) reduces to Eq. (6).

When *d*-band conduction is neglected, the last two terms in the bracket of Eq. (7) vanish and the Hall coefficient is given by an expression similar to Eq. (6) except that conduction in both bands is electronic. If for the conduction electrons the two spin directions are equally probable then the expression for the Hall coefficient reduces to Eq. (3). However, when *d*-band conduction can not be neglected, which seems to be the case for Ni-Pd alloys, the more complete expression given by Eq. (7) must be used.

It is known from magnetic measurements that both Ni and Pd have about 0.6 hole per atom in the *d* band, so that it is reasonable to assume, as was suggested by Wohlfarth,¹⁰ that this number remains unchanged upon alloying. Individually, however, the number of holes per atom in each half of the *d* band will depend upon the atomic fraction x of Pd in Ni. This dependence can be found experimentally from the relative saturation magnetization per atom

$$\zeta_0 = (n_{da} - n_{dp}) / (n_{da} + n_{dp}) = (n_{da} - n_{dp}) / n_s,$$

⁵ F. E. Allison and E. M. Pugh, Phys. Rev. **102**, 1281 (1956).

⁶ F. P. Beitel and E. M. Pugh, Phys. Rev. **112**, 1516 (1958).

⁷ B. R. Coles, Phys. Rev. **101**, 1254 (1956).

⁸ Y. Dorfman and P. Zhukova, Zhur. Eksp. i Teoret. Fiz. **9**, 51 (1939).

⁹ E. H. Sondheimer, Proc. Roy. Soc. (London) **A193**, 484 (1948).

¹⁰ E. P. Wohlfarth, J. Phys. Chem. Solids **1**, 35 (1956).

where $n_s = n_{da} + n_{dp}$ is the number of s -band electrons per atom.

The conductivity of each sub-band may be written¹¹

$$\sigma = (e^2/3\pi^2\hbar^2)k^2(dE/dk)\tau,$$

which is to be evaluated for the appropriate sub-band at the Fermi surface. The relaxation time τ is inversely proportional to the scattering probability P . If we consider only the residual resistivity of an alloy of concentration x , then the scattering probability may be written as¹²

$$P \propto x(1-x)[S(k^2dk/dE)_s + D(k^2dk/dE)_d].$$

Here S is the square matrix element for scattering to the appropriate half of the s band and D the square matrix element for scattering to the appropriate half of the d band. The term (k^2dk/dE) is proportional to the density of final states per atom at the Fermi surface. All terms in the bracket may be taken as independent of x and of spin direction, except for the term $(k^2dk/dE)_d$ which will depend upon both.

Usually it is assumed that the d band forms an inverted band of standard form, in which case the density of states is proportional to the one-third power of the number of holes in the band. The most obvious generalization is to take the density of states proportional to some arbitrary power f of the number of holes in the band. Then the above equations may be combined to give for the effective number of electrons per atom, as defined by Eq. (4),

$$n^* = (n_s/2)(\beta T_1 + T_2)^2 / (\beta^2 T_3 - T_4), \quad (8)$$

where

$$\begin{aligned} T_1 &= 1/(a+u^f) + 1/(a+v^f) = t_1(u) + t_1(v), \\ T_2 &= u^p/(b+u^f) + v^p/(b+v^f) = t_2(u) + t_2(v), \\ T_3 &= t_1^2(u) + t_1^2(v), \\ T_4 &= t_2^2(u)/u + t_2^2(v)/v: \end{aligned}$$

where a , b , and β are constants involving the square matrix elements for scattering, and $u = 1 + \zeta_0$, $v = 1 - \zeta_0$, and $p = \frac{4}{3} - f$. Similarly, the resistivity ρ is given by

$$x(1-x)/\rho = K(\beta T_1 + T_2). \quad (9)$$

The right-hand sides of Eqs. (8) and (9) depend on x only through $u = 1 + \zeta_0$ and $v = 1 - \zeta_0$.

This last Eq. (9) was used by Overhauser and Schindler¹³ in their discussion of the residual resistivity of Ni-Pd alloys. They were able to obtain a good fit to their data even when they neglected the term representing d -band conduction. We retain this term here, however, since it is crucial to an explanation of the Hall effect in these alloys.

¹¹ N. F. Mott and H. Jones, *Theory of the Properties of Metals and Alloys* (Oxford University Press, Oxford, 1936).

¹² A. H. Wilson, *The Theory of Metals* (Cambridge University Press, New York, 1953).

¹³ A. W. Overhauser and A. I. Schindler, *J. Appl. Phys.* **28**, 544 (1957).

EXPERIMENTAL METHOD

The samples used in this investigation were prepared by melting under vacuum in a pure Al_2O_3 crucible. The resulting ingots were rolled to a thickness of about $\frac{1}{16}$ inch, and samples, probes and wire made from these sheets. After machining, the samples were annealed in vacuum for a period of 12 hours at 650°C. The samples were in the form of flat plates nominally 1 cm wide, 10 cm long, and 1 mm thick. Four probes of the same material as the sample were made for each. Two of these were placed on opposite sides of the sample for measuring the Hall effect and the other two on the same side of the sample and about 4 cm apart for measuring the resistivity. Pieces of alloy wire were soldered to each probe and the junctions of the alloy wires and copper leads were placed inside a copper box to eliminate thermal emf's and any errors due to the Ettingshausen and Righi-Leduc effects. Two thermocouples were clamped independently to the ends of the sample. The samples and probes were rigidly held in a holder designed for this purpose, and the probes held in contact with the edges of the sample by means of phosphor bronze springs.

The temperature dependence of the Hall effect was determined by measurements in the temperature range from 4°K to room temperature. The baths used, which were in direct contact with the sample, were liquid helium (4°K), liquid hydrogen (20°K), liquid methane (112°K), liquid ethylene (169°K), and a silicone oil with high thermal conductivity (room temperature).

The reversal in sign of the Hall potential with magnetic field was offset by introducing a bias voltage into one Hall potential lead. The total potential was measured with a Rubicon thermofree potentiometer and a Rubicon photoelectric galvanometer amplifier. The incremental method³ was used to measure the Hall potential for fields up to 3.1 webers/m².

DATA ANALYSIS

For a flat plate, Eq. (1) can be written

$$Vt/I = R_0B + (R_1 - R_0)M. \quad (10)$$

The derivative with respect to B of this equation is denoted by R_0^* and is given by

$$R_0^* = R_0 + (R_1 - R_0)\partial M/\partial B. \quad (11)$$

For the samples and temperatures used here the second term is negligible in the region where B is sufficient to saturate the sample, so that no distinction need be made between R_0 and R_0^* . Thus, R_0 and R_1 could be obtained directly by fitting a least squares straight line of the form

$$Vt/I = R_0B + (R_1 - R_0)M_s, \quad (12)$$

to the high field points, where M_s is the saturation magnetization of the sample. In every case it was found that the data obeyed very well a relation of this form for B greater than about 1.6 webers/m².

Attempts were also made to separate the Hall and Nernst effects by the method of Allison and Pugh,⁵ but no correlation could be found between the longitudinal temperature gradient and the measured values of R_0 and R_1 . The longer samples used in this investigation were apparently sufficient to eliminate this possible source of error in the measured values of the Hall coefficients.

EXPERIMENTAL RESULTS

Measurements were made on Ni-Pd alloys containing 2, 5, 10, 20, 30, and 60 atomic percent Pd. In addition, measurements were made on a Ni sample of 99.99% purity furnished by the Office of Naval Research. Curves of the ordinary Hall coefficient as a function of temperature and composition are shown in Fig. 1. Particularly interesting in these curves is the crossover at low temperatures between the curve for pure Ni and that for the most dilute alloy. Also interesting is the relatively

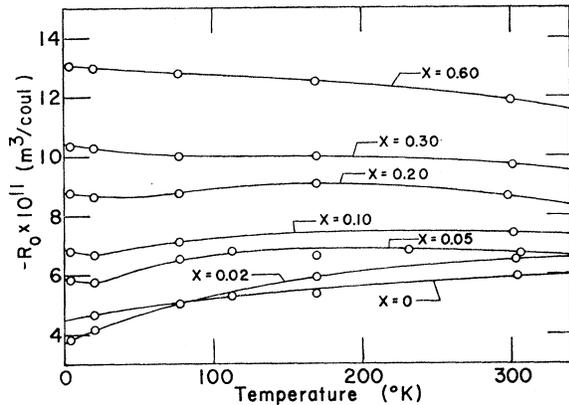


FIG. 1. The ordinary Hall coefficient for Ni-Pd alloys. The numbers attached to the curves give the atomic fraction of Pd in Ni.

weak dependence of the ordinary Hall coefficient on impurity content. These points are illustrated by Fig. 2, which compares the variation of R_0 with impurity content for Ni-Pd¹⁴ and Ni-Cu¹⁵ alloys at 20°K.

The measured values of the residual resistivity (4°K) are shown in Fig. 3. The figure includes also the data of Overhauser and Schindler,¹³ who discussed the significance of the curve. Our data are in good agreement with theirs.

DISCUSSION

Ordinary Effect

According to the theoretical considerations discussed above, the variation of both the ordinary Hall coefficient and the residual resistivity in these alloys depends only

¹⁴ The point shown for pure Pd is from some unpublished work of F. E. Allison and B. R. Coles. We are grateful to them for permission to quote this value.

¹⁵ P. Cohen, Office of Naval Research Technical Report, June 1955 (unpublished); thesis, Carnegie Tech, 1955 (unpublished).

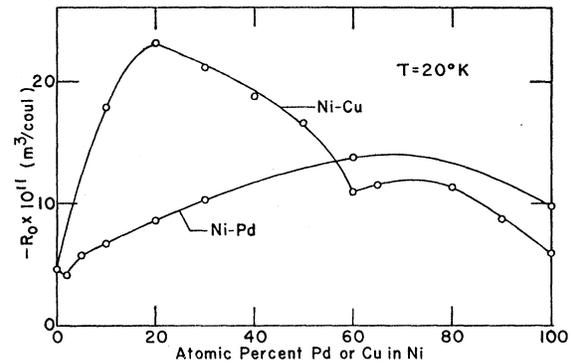


FIG. 2. The ordinary Hall coefficient at $T=20^\circ\text{K}$ for Ni-Pd and Ni-Cu alloys. The curve for the Ni-Cu alloys shows R_0^* , not R_0 .

upon the number of electrons in the two halves of the d band, which can presumably be found from saturation magnetization measurements. This dependence is given in Eqs. (8) and (9), which include some parameters whose values are unknown and cannot be independently found. To determine the values of these parameters we must, for example, choose them so that Eq. (8) best describes the measured Hall coefficients. Then the residual resistivity can be directly calculated from Eq. (9) and the values so obtained compared with the measured ones. Because of the incompleteness of the magnetic information the procedure just outlined had to be modified.

We are concerned mainly with the variation of n^* with Pd content in Ni at the absolute zero of temperature. This should be given as a function of the relative saturation magnetization of these alloys by Eq. (8). If the variation of ζ_0 with x , the atomic fraction of Pd in Ni, is known from magnetic measurements, then the various constants in this equation can be found by fitting the equation to the measured values of n^* . But ζ_0 can be found from the magnetic measurements only if its value is known for at least one value of x . It is

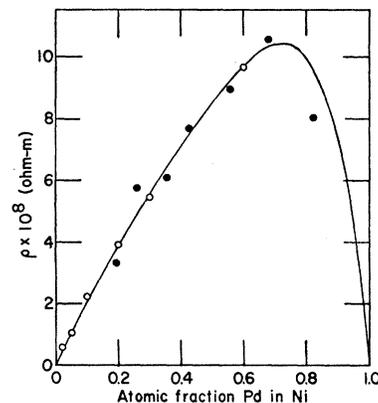


FIG. 3. The residual resistivity of Ni-Pd alloys as a function of composition. The solid circles are taken from the work of Overhauser and Schindler.

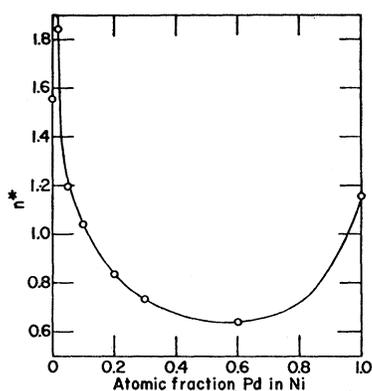


FIG. 4. The effective number of electrons per atom for Ni-Pd alloys at $T=4^{\circ}\text{K}$. The curve shown was calculated using Eq. (8).

usually assumed on the basis of a theoretical treatment of the high-temperature magnetic susceptibility of Ni¹⁶ and on the basis of the variation of the saturation magnetization of Ni-Cu alloys that $\zeta_0=1$ for Ni. But this value is only approximate, and for our purposes ζ_0 must be known quite accurately, particularly when it is close to unity. Thus, it is not practical to try to fit Eq. (8) to the data directly. Instead, we have assumed that the residual resistivity, as given by the curve shown in Fig. 3, can be described by Eq. (9). Also, since the magnetic measurements indicate¹⁰ that ferromagnetism just vanishes for 97% Pd in Ni, it was assumed that $\zeta_0=0$ for $x=0.97$ and that for this value of x , $n^*=1.037$ at $T=0^{\circ}\text{K}$. These assumptions determine everything except one of the parameters in Eqs. (8) and (9), which can then be used to attempt a fit to the curve of n^* as a function of x . A separate verification can then be obtained by comparing the deduced values of ζ_0 with the measured saturation magnetizations of these alloys, since now ζ_0 is known accurately for pure Ni.

The results of the calculations are shown in Fig. 4, which compares the measured values of n^* at $T=4^{\circ}\text{K}$ and a curve calculated from Eq. (8) using the values

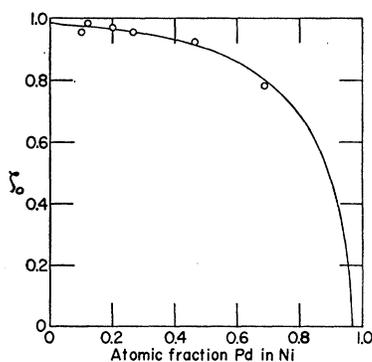


FIG. 5. The relative saturation magnetization of Ni-Pd alloys at $T=0^{\circ}\text{K}$. The curve shown was deduced from the measured values of the ordinary Hall coefficient and the residual resistivity.

¹⁶ E. P. Wohlfarth, Proc. Roy. Soc. (London) **A195**, 434 (1949).

$f=1$, $a=0.09006$, $b=0.044$, $\beta=3.957$, and values of ζ_0 deduced from the residual resistivity curve shown in Fig. 3. For n_s we used the value 0.604, which may be somewhat too large. The agreement between the theoretical curve and the data is excellent, but it should also be mentioned that relatively small changes in the residual resistivity curve shown in Fig. 3 can result in relatively large changes in Fig. 4, at small values of x . The disagreement for pure Ni is of some significance. We feel this indicates that the assumption of a relaxation time that is a function of the energy only is not a valid assumption for thermal scattering in these alloys, although it is for impurity scattering. This is in agreement with a naive picture of the processes involved.

Figure 5 shows the deduced curve of ζ_0 as a function of x , as compared with values calculated from the saturation magnetization measurements of Sadron¹⁷ and

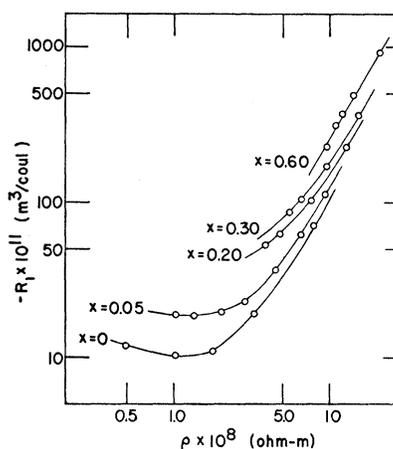


FIG. 6. Extraordinary Hall coefficients for Ni-Pd alloys plotted against the resistivity on a log log scale. The curves connect values measured for the same sample at different temperatures. Numbers attached to the curves give the atomic fraction of Pd in Ni.

Went.¹⁸ The points shown were calculated from the measurements by

$$\zeta_0 = \zeta_{00}(1 + 0.818x)\sigma_0(x)/\sigma_0(0),$$

where x is the atomic fraction of Pd in Ni and $\sigma_0(x)$ is the saturation moment per gram at $T=0^{\circ}\text{K}$ for an alloy of concentration x . For ζ_{00} , the relative saturation magnetization for pure Ni, we used the value 0.985 which was necessary to fit the curve of n^* versus x . The agreement between the deduced curve and the measured values is very good, and we feel that it offers the most convincing argument in support of our explanation of the ordinary Hall effect in these alloys.

Extraordinary Effect

Until recent years, the extraordinary Hall coefficient in ferromagnetic materials has received much less at-

¹⁷ C. Sadron, Ann. phys. **17**, 371 (1932).

¹⁸ J. J. Went, Physica **17**, 98 (1951).

tion than has the ordinary Hall coefficient. This was due to the fact that an adequate theory has been much more difficult to find for R_1 than it has for R_0 .

Recently, Karplus and Luttinger¹⁹ derived a relation between the extraordinary Hall coefficient R_1 and the resistivity ρ on the basis of spin-orbit interaction. Similar results have since been obtained by a number of other authors.²⁰⁻²² The various theoretical relationships proposed in these papers have been tested by many investigations. The extraordinary Hall coefficient and the resistivity have been varied by varying the temperature,^{5,23} the degree of order,^{24,25} and the impurity content.^{6,20} The degree of agreement between theory and experiment has varied greatly from one material to another.

The alloy system considered here should be quite useful in distinguishing between the various theoretical relationships. In Fig. 6 are shown the results for R_1 on the usual $\log R_1$ versus $\log \rho$ plot. The various curves shown in the figure connect values measured for the same sample at different temperatures. The results are presented in a different form in Fig. 7 where the curves shown connect values obtained for different samples at the same temperature.

For high temperatures and high impurity content, the curves shown in Fig. 6 and Fig. 7 all appear to approach a relation of the form

$$R_1 = a\rho^n. \quad (13)$$

The coefficient a seems to vary in a regular manner with temperature and impurity content. For the curves shown in Fig. 6 the exponent $n=1.55$, and for the curves shown in Fig. 7, $n=2.5$. This latter is the only case of which we know where n exceeds 2.

On the whole, however, the results shown in Fig. 6 and Fig. 7 can support neither a relation of the form Eq. (13) nor one of the form

$$R_1 - R_0 = b\rho^n, \quad (14)$$

particularly not if $n=2$ as is required by the theories.

¹⁹ R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).

²⁰ J. Smit, Physica **21**, 877 (1955).

²¹ J. M. Luttinger, Phys. Rev. **112**, 739 (1958).

²² P. N. Argyres, Phys. Rev. **97**, 334 (1955).

²³ C. Kooi, Phys. Rev. **95**, 843 (1954).

²⁴ S. Foner, F. E. Allison, and E. M. Pugh, Phys. Rev. **109**, 1129 (1958).

²⁵ J. P. Jan, Helv. Phys. Acta **30**, 233 (1957).

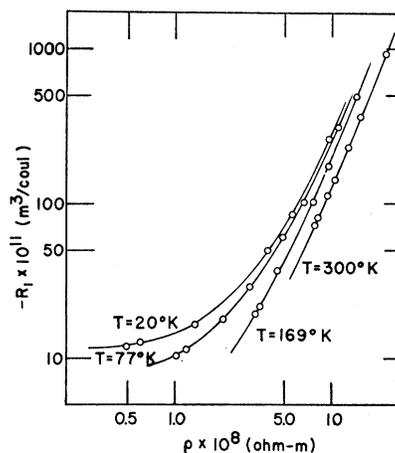


FIG. 7. Extraordinary Hall coefficients for Ni-Pd alloys plotted against the resistivity on a log log scale. The curves connect values measured for samples of differing Pd content at the indicated temperature.

Furthermore, we have also been unsuccessful in representing the data for $T=4^\circ\text{K}$ by the more general relation

$$R_1 = bR_0 + c\rho + a\rho^2, \quad (15)$$

suggested by the paper of Luttinger.²¹

The data show also the deviation from a simple power law at low temperatures that has been found in so much of the previous work. This deviation is usually attributed to either the term bR_0 ¹⁹ or to impurities,²⁰ but neither explanation seems to adequately describe the data. Also, it has been found^{3,6} in some alloy systems that both R_1 and $R_1 - R_0$ change sign as the temperature or impurity content is varied. In view of these facts, it is necessary to conclude that there is some additional factor, not included in the existing theories, that is necessary to completely describe the extraordinary Hall effect.

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