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Hall Effects, Resistivity, and Thermopower in Fe and $Fe_{1-x}Ni_x$ for $x=0$ to 0.2*

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Measurements were made of the ordinary and extraordinary Hall effects and the electrical resistivity in a series of iron-nickel alloys from pure iron to 20% nickel in iron, in the temperature range from 77°K to room temperature. The thermopower of the same alloys was measured in the range from 200 to 300°K. The ordinary Hall, thermopower, and resistivity data indicate that hole conduction is important for pure iron, and that as nickel is added electron conduction becomes more important. The location and shape of the Fermi surface is estimated from the results of an energy-band calculation, using a rigid-band model. The results are in agreement with increasing electron and decreasing hole contributions as nickel is added to iron. Both the data and the energy-band results seem to lead to a complicated multiband picture at low nickel concentrations and an approach towards single-, nearly spherical-, band conduction at 20% nickel. The temperature and composition dependence of the ordinary Hall coefficients are accounted for qualitatively by estimates of the anisotropy of the relaxation time and of the effect of impurity scattering on the relaxation time. The extraordinary Hall coefficient is fitted by a number of models based upon theories by various authors. Fairly good agreement with experiment is obtained over a wide range of composition and temperature, but with no clear preference for any of the theories.

INTRODUCTION

It has been well established that in ferromagnetic materials the Hall electric field per unit current density ρ_H is given by

$$\rho_H = R_0 B + R_s M, \tag{1}$$

where B is the magnetic induction and M the magnetization in the material, and where R_0 and R_s are the ordinary and spontaneous Hall coefficients, respectively.[1] A model for the ordinary Hall coefficients of metals and alloys of the iron transition series has been proposed by Pugh.[2] This model is based upon a mechanism originally suggested by Mott[3] to explain the electrical resistivity of transition elements. The central idea of the model is that the d-band electrons of spin up and spin down in ferromagnetic materials should not be considered as being in a single band, since they have different occupation numbers. Also, since s-d transitions are more likely to occur between states of the same spin, the s-band electrons should likewise be divided into spin-up and spin-down sub-bands. Then the s-to-d transition rate will depend on the density of states in the d sub-band of like spin. Applications of this model have generally assumed a spherical Fermi surface (not necessarily parabolic) and isotropic relaxation times, but in spite of these simplifying assumptions the model has been successful in explaining the experimentally observed ordinary Hall coefficients of a number of alloys in this series.[4–8] The results of applying this model to ternary alloys of Cu, Ni, and Fe,[7,8] for example, were consistent with the assumption that the 4s electrons carried most of the current, unless the alloys had a low impurity content. For the pure metals, however, the ordinary Hall coefficient R_0 generally shows an upward cusp when plotted against electron concentration. There the assumption of the 4s electrons being predominant, and the assumptions of a simple spherical band with isotropic relaxation times, appear to break down.

Hall-effect measurements in some alloys of the Fe-Co system have been made by Beitel and Pugh.[5] Their measurements for electron numbers near iron were carried out on alloys containing from 0.1 to 35 at. % Co, and at temperatures between 77°K and room temperature. The measurements showed a sharp increase (decrease in absolute value) as the cobalt content is decreased, with R_0 becoming positive in pure iron. The data were analyzed in terms of n^*, an effective number of electrons per atom, defined by

$$n^* = -1/R_0 N e, \tag{2}$$

where N is the number of atoms per m^3 and e is the magnitude of the electronic charge in coulombs. This quantity, which is introduced by analogy with the corresponding equation for a simple single-band conductor, was found for these Fe-Co alloys to increase from about 0.3 at 35% Co to 0.6 at 15% Co and then to very large or to negative values at 0.5% and 0.1% Co. There, as is the case near pure Ni, an expression of the form (2) is not very meaningful, since more than one kind of carrier contributes importantly to R_0.

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[1] A third coefficient, usually denoted by R_1 and called the extraordinary coefficient, is also often used. It is related to the coefficients of Eq. (1) by $R_1 = R_0 + R_s$.

[2] E. M. Pugh, Phys. Rev. **97**, 647 (1955).

[3] N. F. Mott, Proc. Roy. Soc. (London) **A153**, 699 (1936).

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

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

[6] J. A. Dreesen and E. M. Pugh, Phys. Rev. **120**, 1218 (1960).

[7] E. R. Sanford, A. C. Ehrlich, and E. M. Pugh, Phys. Rev. **123**, 1947 (1961).

[8] A. C. Ehrlich, J. A. Dreesen, and E. M. Pugh, Phys. Rev. **133**, A407 (1964).

In order to investigate further the behavior of R_0 in the region of pure iron, we have carried out Hall-effect and other measurements on a sample of high-purity iron and on some iron-rich alloys of the Fe-Ni system. It was hoped that some additional details of the dependence of R_0 on electron concentration in the region of pure iron would be found. In particular, the region is one in which the 3*d* band is likely to be an important contributor and in a way that is not likely to be described by spherical Fermi surfaces and isotropic relaxation times. The investigation was also designed to show the effect on R_0 of different impurities at the same electron numbers in iron alloys.

EXPERIMENTAL METHODS

Samples for Hall-effect and resistivity measurements were made in the form of rectangular plates approximately 10 cm long, 1 cm wide, and 1 mm thick. A pure iron sample was made from an ingot of zone-refined iron which contained less than 185 ppm of analyzed metallic impurities. The other samples contained nominal percentages of Ni to the amounts of 0.4, 5, 10, 15, and 20 at. %. Ingots of the desired compositions were prepared by melting in a vacuum induction furnace, and after rolling, samples, probes, and wire were made from each ingot. All samples and probes were subsequently annealed to minimize strains. Cold rolling was used for the pure iron ingot and for the ingots containing 0.4, 5, and 10% Ni, while hot rolling was found to be necessary for the ingots containing 15 and 20% Ni. Phase diagrams of the Fe-Ni system indicate that a mixed fcc and bcc phase occurs at room temperature and below for alloys containing more than about 15 or 20% Ni. The boundary of this mixed-phase region is not well established, but our inability to cold roll both the 15 and 20% samples and the ease with which the others could be cold rolled seem to indicate that both our 15 and 20% samples were of mixed phases in the temperature range studied.

The sample and probes were held in place by a Micarta sample holder, with phosphor bronze springs used to ensure electrical contact between probes and sample. Two thick copper sheets, electrically insulated from the sample by a dielectric spray coat and a thin Mylar sheet, were placed on the sample in an effort to minimize any temperature gradient. Resistivity and Hall measuring circuits were set up with the alloy-to-copper wire junctions enclosed in copper cylinders. The junctions in each circuit were immediately adjacent to one another and in as good thermal contact as possible. In this way thermal emfs were reduced to a minimum and possible errors due to the Ettingshausen effect eliminated. In the case of the resistivity circuit, the copper wires led to a Wenner potentiometer and a dc null detector. For the Hall-effect measurements a Rubicon Thermofree potentiometer and a photoelectric amplifier were used. This latter circuit was capable of measuring differences of 10^{-9} V, although the accuracy was probably not this good. In addition, thermocouples were placed at each end of the sample and were used for monitoring the sample temperature.

The sample temperature was stabilized by placing the sample in baths of boiling nitrogen (77°K), methane (112°K), ethylene (169°K), propane (231°K), and in some cases boiling argon (87°K). For room-temperature measurements a bath of silicone oil having a high thermal conductivity was used. A special two-walled Dewar described by Beitel and Pugh[5] was used to contain the baths and to increase temperature stability.

The magnetic fields were produced by an A. D. Little electromagnet whose current was controlled by an electronic feedback control circuit. A rotating coil fluxmeter was used to measure the magnetic fields and to obtain a calibration curve of magnetic field versus magnet current. The largest field obtainable with the experimental setup was approximately 3.3 Wb/m^2 (33 000 G) and the fields were measured to about 0.1%.

The absolute thermopower of iron of the Fe-Ni alloys containing 5, 10, 15, and 20 at. % Ni were measured in the temperature range from 200 to 300°K. The baths used for this purpose were acetone-cooled with dry ice and liquid propane cooled below its boiling point by liquid nitrogen. The bath temperature was allowed to drift and the emf of the sample material versus copper and the bath temperature alternately measured. The measured thermal emf's were converted to absolute thermopowers using the data for copper given by Borelius.[9] The results showed considerable scatter and the temperature dependence of the thermopower could not be determined accurately. However, the composition dependence was readily determined.

DATA ANALYSIS

The Hall coefficients are determined from the relation (1), which in terms of experimentally measured quantities may be written as

$$\rho_H = Vt/I = R_0B + R_sM\,, \tag{3}$$

where V is the transverse Hall potential, t the sample thickness, and I the primary sample current. Above magnetic saturation of the sample the second term becomes constant and when ρ_H is plotted against B a straight line results. The slope of this straight line gives the ordinary Hall coefficient R_0 and if the line is extended backwards to $B=0$ the intersection with this axis gives the quantity R_sM_s, where M_s is the saturation magnetization of the sample. The slope and the intersection of this straight line were determined by a least-squares fit to the data well above magnetic saturation, as indicated by ρ_H versus B being linear within the experimental scatter of the data. The spontaneous

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[9] G. Borelius, *Handbuch der Metallphysik*, edited by G. Masing (Akademische Verlagsgesellschaft m.b.h., Leipzig, 1935), p. 398.

Hall coefficient was then determined from the intercept by dividing its value by the saturation magnetization.[10]

To allow for the possibility of incomplete magnetic saturation, this high-field slope of ρ_H versus B is generally denoted by R_0^*, where

$$R_0^* = R_0 + R_s(dM/dB). \qquad (4)$$

Study of the existing magnetization data for this alloy system[10] shows that R_0 is within 3×10^{-11} m³/C of R_0^*, and although in some cases this difference may amount to a large fraction of R_0, no distinction will be made in the following between R_0 and R_0^*. Similarly, the spontaneous Hall coefficient R_s will be considered the same as R_s^* which is based upon R_0^* instead of R_0.

EXPERIMENTAL RESULTS

The measured values of R_0, R_s, and the electrical resistivity are shown for each composition as functions of the temperature in Figs. 1, 2, and 3. Figure 1 includes also a scale giving n^*, the effective number of electrons per atom, as calculated by Eq. (2). The factor relating n^* to R_0 is, strictly speaking, both temperature and composition dependent, but for the ranges of temperature and composition studied here it is sufficiently constant that a single scale for n^* can be used for all the data shown in Fig. 1. R_0 and the absolute thermopower Q, both at 280°K, are shown as functions of compositions in Fig. 4.

DISCUSSION

Ordinary Hall Coefficients and Thermopower

One of the most striking features of the data is the similarity of the ordinary Hall coefficient R_0 and the thermopower Q near room temperature, as shown in Fig. 4. Both are positive for pure iron, change sign between pure iron and 5% Ni, and continue to decrease as more Ni is added. If the diffusion term of the thermopower is assumed to be dominant at these temperatures, then the behavior of both R_0 and Q is consistent with the view that conduction is predominantly by holes in pure iron, with the electron contribution increasing and/or hole contribution decreasing as Ni is added.[11] This could occur, for example, if in pure iron there are hole pockets in the corners of a given Brillouin zone and electron pockets in higher zones. Then, since the Fermi energy is raised as Ni is added to iron, the volume occupied by electrons increases as the Ni content is increased. An exception could occur, however, if portions of the Fermi surface surrounding electron pockets started to contact a zone boundary as Ni is added. The overlap would then result in a replacement of some of the electron states by hole states. The effect of this on R_0 and Q would be a tendency for these to increase as more Ni is added. The data shown in Fig. 4 do not exhibit such a behavior and are consistent with a lack of such a zone boundary contact in this range of compositions.

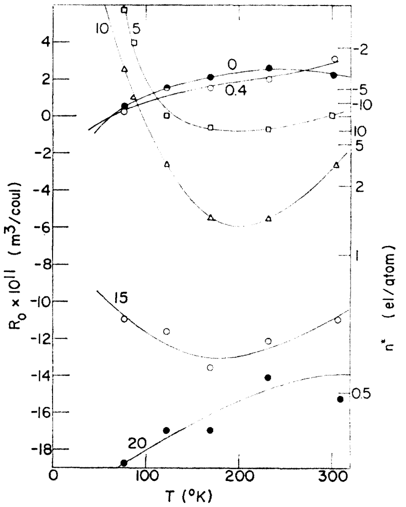

FIG. 1. The ordinary Hall coefficient and effective number of electrons per atom as functions of temperature for iron-nickel alloys. The numbers associated with the curves give the atomic percent nickel in iron.

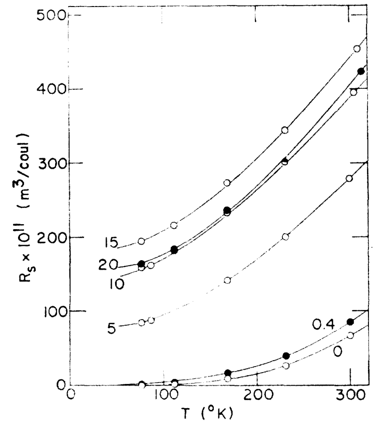

FIG. 2. The spontaneous Hall coefficient as a function of temperature for iron-nickel alloys. The numbers associated with the curves give the atomic percent nickel in iron.

The behavior described above can be justified qualitatively by making use of the energy-band calculation of Wood[12] for bcc iron. We have used Wood's calculations without considering the effect of spin other than to place the electrons of opposite spin in two sub-bands of the same form except for an adjustment of the Fermi level relative to the bottom of these sub-bands. The shift was such that the difference between occupation

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[10] The spin-wave model $M_s(T)=M_s(0)\ (1-CT^{3/2})$ was used as an interpolation formula, $M_s(0)$ and C being obtained from the work of M. Peschard, Compt. Rend. **180**, 1836 (1925).

[11] J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, England, 1962), p. 397. The diffusion term of the thermopower is assumed dominant at these temperatures. In the present discussion, portions of the Fermi surface surrounding occupied (unoccupied) states in the reduced-repeated zone scheme are considered to contribute electron (hole) states. The sign with which these states contribute to R_0 and Q depends upon the effective mass tensor. For the most part, however, the holes (electrons) should contribute positive (negative) terms to both.

[12] J. H. Wood, Phys. Rev. **126**, 517 (1962).

numbers below the Fermi energy gives the correct magnetization, after correction for orbital contribution, of iron. For the alloys we have assumed a rigid-band model so that the only effect of adding Ni to iron is to alter the Fermi levels of each sub-band without changing the shapes or positions of the constant-energy surfaces in wave-vector space. The change in the Fermi energy ζ upon alloying was taken so that the total changes in occupation numbers are given by

$$n_{\pm}(x)-n_{\pm}(0)=\int_{\zeta_{\pm}(0)}^{\zeta_{\pm}(x)}\mathfrak{N}_{\pm}(\mathcal{E})d\mathcal{E}. \qquad (5)$$

Here x is the atomic fraction of nickel in iron and the subscripts $\pm$ refer to the parallel and antiparallel spin sub-bands, where all electrons of a given spin are included in each. The Fermi energies $\zeta_{\pm}(x)$ are measured relative to the bottom of each spin sub-band and $\mathfrak{N}_{\pm}(\mathcal{E})$ are the densities of states. The values of $n_{\pm}(x)$ were calculated from the conditions

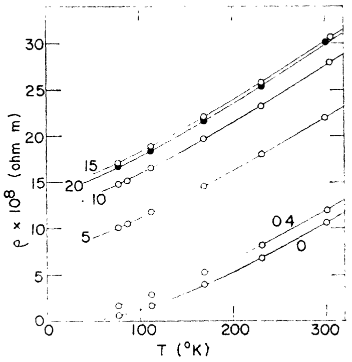

FIG. 3. The resistivity as a function of temperature for iron-nickel alloys. The numbers associated with the curves give the atomic percent nickel in iron.

$$n_{+}(x)+n_{-}(x)-n_{+}(0)-n_{-}(0)=2x \qquad (6)$$

and the approximate difference

$$n_{+}(x)-n_{-}(x)=\tfrac{1}{2}g'(x)m_s(x)\,, \qquad (7)$$

where $m_s(x)$ is the magnetic moment per atom in Bohr magnetons and $g'(x)$ is the magnetomechanically determined gyromagnetic ratio.[13] The shapes of the various parts of the Fermi surface were constructed graphically, and it was found that they could be described approximately, but concisely, by expansions to the first Kubic harmonic Y_c^4 of the wave vector terminating on the Fermi surface,

$$k=k_0+k_1Y_c^4(\theta,\phi)\,,$$

where θ and ϕ are the polar angles of k and where k_0 and k_1 are constants which depend on the Fermi energy. A similar procedure has been used by Davis[14] and by Cooper and Raimes.[15]

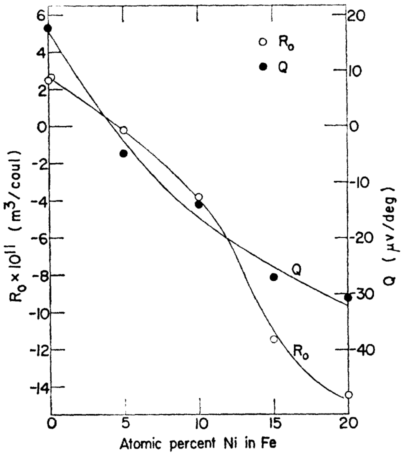

FIG. 4. The ordinary Hall coefficient and the absolute thermopower at $T=280$°K, as functions of composition for iron-nickel alloys.

The results of the calculations may be summarized as follows: The pockets enclosing occupied and unoccupied states can be referred to by the notation $(m)nS_{\pm}$, where n is the band number of Wood,[12] S is the symmetry element in the center of the band in the reduced repeated zone scheme, using the notation of Bouckaert, Smoluchowski, and Wigner,[16] and m is the number of bands per atom per cubic symmetry center when different from unity. The values of the coefficients k_0 and k_1, as determined from Wood's results, are listed in Table I. The small $(6)3N_{-}$ and $(6)5\Delta_{-}$ pockets, not at symmetry points, were assumed spherical for simplicity.

The results of Table I show the emergence of some small electron pockets as Ni is added. However, cross sections of all these pockets in wave-vector space show that there is no tendency for the onset of overlap of the zone boundaries, as Ni is added, by pockets enclosing occupied states at iron, with the possible exception of the highly nonspherical $4\Gamma_{-}$ pocket barely touching at about 20% Ni in Fe. That is, the topology of the pockets does not appear to make any major changes, by the assumed model, in the range between Fe and 20Ni-80Fe. The pocket which should contribute the largest term to R_0 is the $6\Gamma_{+}$, which by itself could give $n^*=0.3$ or $R_0=-24$. This is fairly close to the range of measured values of R_0 in $\mathrm{Ni}_{0.20}\,\mathrm{Fe}_{0.80}$.

The temperature and composition dependence of R_0 at the lower temperatures is not easily explained quantitatively. Also, whereas both the ordinary Hall and thermopower data at higher temperatures are consistent with the simple band model described above, at lower temperatures the variations in the relaxation time may be more important. A possible explanation of the lower temperature behavior can be derived by noting that there are more holes than electron pockets, but that

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[13] G. Scott, Rev. Mod. Phys. **34**, 102 (1962).

[14] L. Davis, Phys. Rev. **56**, 93 (1939).

[15] J. Cooper and S. Raimes, Phil. Mag. **4**, 145 (1959).

[16] L. P. Bouckaert, R. Smoluchowski, and E. Wigner, Phys. Rev. **50**, 58 (1936).

Table I. Energies and band parameters for Fe-Ni alloys, as determined from Wood's energy-band calculations.[a]

Atomic % Ni in Fe	Pocket	ζ_+ (Ry)	ζ_- (Ry)	k_0 $(\pi/4a)$	k_1 $(\pi/4a)$
0	$6\Gamma_+$	0.830		3.9	−0.039
	$4\Gamma_-$		0.690	1.8	1.7
	$3H_-$		−0.690	2.4	0.65
	$5H_+$	−0.830		2.9	−0.97
	$3H_+$	−0.830		0.36	0.014
	$4H_+$	−0.830		1.5	−0.17
	(6) $3N_-$		−0.69	0.94	0
	(6) $5\Delta_-$		−0.69	0	0
5	$6\Gamma_+$	0.835		4.0	−0.034
	$4\Gamma_-$		0.692	1.9	1.8
	$3H_-$		−0.692	2.4	0.65
	$5H_+$	−0.835		2.5	−1.0
	$3H_+$	−0.835		0.27	0.075
	$4H_+$	−0.835		1.3	−0.12
	(6) $3N_-$		−0.692	0.93	0
	(6) $5\Delta_-$		0.692	0	0
10	$6\Gamma_+$	0.838		4.0	−0.031
	$4\Gamma_-$		0.697	2.0	2.0
	$3H_-$		−0.697	2.3	0.63
	$5H_+$	−0.838		2.2	−1.0
	$3H_+$	−0.838		0.21	0.0051
	$4H_+$	−0.838		1.2	−0.099
	(6) $3N_-$		−0.679	0.91	0
	(6) $5\Delta_-$		0.697	0	0
15	$6\Gamma_+$	0.841		4.1	−0.025
	$4\Gamma_-$		0.703	2.3	2.2
	$3H_-$		−0.703	2.2	0.60
	$5H_+$	−0.841		2.0	−1.1
	$3H_+$	−0.841		0.16	0.004
	$4H_+$	−0.841		1.0	−0.072
	(6) $3N_-$		−0.703	0.88	0
	(6) $5\Delta_-$		0.703	0.90	0
20	$6\Gamma_+$	0.844		4.2	−0.027
	$4\Gamma_-$		0.708	2.5	2.4
	$3H_-$		−0.708	2.1	0.57
	$5H_+$	−0.844		1.6	−1.1
	$3H_+$	−0.844		0.061	0.004
	$4H_+$	−0.844		0.79	−0.048
	(6) $3N_-$		−0.708	0.85	0
	(6) $5\Delta_-$		−0.708	0.38	0

[a] The notation for labeling pockets is this: The letter gives the center, the sign gives the spin, the number preceding the letter gives the band number in Wood's notation, and the number in parentheses gives the number of pockets per atom when different from one. A positive value for the Fermi energy indicates that the surface surrounds occupied states and a negative value that the surface surrounds unoccupied states.

the *average* volume enclosed by the hole pockets is less than that enclosed by electron pockets (excluding the very small (6)$5\Delta_-$ pockets). Recent magnetoresistance data obtained at high fields by Fawcett and Reed[17] indicate that iron is a compensated metal, i.e., has equal numbers of occupied and unoccupied states in its pockets. Thus, a larger number of hole pockets should lead to a smaller average-hole-pocket size. At a given temperature, scattering of electrons by phonons in N processes occurs mostly for phonon wave vectors up to a certain length, this length increasing with the absolute temperature. Thus the maximum scattering angle by N processes at a given temperature is larger in pockets of smaller Fermi radii. If the thermal and impurity scattering are independent mechanisms the relaxation time is given by

$$(1/\tau)=(1/\tau^i)+(1/\tau^t) \tag{8}$$

for a given pocket, where $(1/\tau^i)$ and $(1/\tau^t)$ are the impurity and thermal scattering rates, respectively. At 300°K the thermal scattering rate may be large enough so that the relaxation time is almost independent of the impurity scattering rate. Then its composition dependence would be largely due to changes in band properties at the Fermi surface, such as the density of states. However, at 77°K, the effect of the impurity scattering rate on the relaxation time is more important. In particular, changes of the impurity scattering rate should more strongly affect those pockets which are least strongly thermally scattered. By the above argument, these would be the larger or electron pockets, if N processes provide the main thermal scattering mechanism, which is probable at these lower temperatures. This would explain the initial rise in R_0 with Ni content at 77°K. At the higher Ni contents, the decreasing size of hole pockets could cause the decrease in R_0 with Ni content, as at higher temperatures. Also the possible existence of mixed phases at higher Ni content could decrease impurity scattering as discussed below in analyzing the resistivity data.

The sharp decrease in R_0 with temperature at low temperatures in the 5 and 10% samples could be due to the more rapid increase of maximum scattering angle in the smaller hole pockets. If this increase were to "saturate" at 180°, the further increase of angle in the larger electron pockets would be consistent with the subsequent upturn of R_0 versus temperature at higher temperatures.

In the pure and nearly pure (0.4% Ni) iron samples the temperature dependence of R_0 is less marked and the opposite of that expected by the above argument. Since impurity scattering is absent or very small in these samples, it is likely that the anisotropy of the relaxation time is greater. The general trend of the variation of the anisotropy of the relaxation time with temperature, expected in N processes, may be described as follows.

At higher temperatures and large scattering angles (which contribute most to the scattering rate) a wide range of final states may occur and there should be very little dependence on the initial state and hence small anisotropy. As the temperature is decreased somewhat, the allowed range of angles is more dependent on the magnitude of the initial wave vector k_i. It is given approximately at smaller angles by $\theta_m=2\sin^{-1}(2q_m/k_i)$, where θ_m and q_m are the corresponding maximum scattering angles and phonon wave vectors. Here the variation of k over small angles has been neglected. Thus, the relaxation time tends to have an anisotropy similar to the deviation of the Fermi surface from a sphere and of like sign. However, as the temperature and scattering

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[17] E. Fawcett and W. A. Reed, Phys. Rev. **131**, 2463 (1963).

angles become still lower, the effect of the density of final states, which now lie close to the initial states, becomes more important. The density of states (per atom) is given by

$$\mathfrak{N}(\mathbf{k},\mathcal{E})=\frac{1}{(2\pi)^3}\frac{d\omega}{\Omega_{\mathrm{at}}}k^2\frac{dk}{d\mathcal{E}} \tag{9}$$

for one spin and an element of volume in k space, where $d\omega$ is an element of solid angle, Ω_{at} the atomic volume, and $\mathcal{E}$ is the energy, here evaluated at the Fermi surface. Owing to its appearance as a squared quantity k^2 may be more important, and this is approximately k_i^2 at low temperatures. Here the effect of variation in k_i has the opposite effect because of the proportionality between scattering rate and density of final states. Thus the general trend appears to be small anisotropy in thermal relaxation time at high and low temperatures with a maximum in magnitude at intermediate temperatures. Also, if the relaxation time is expanded in a manner analogous to k in Eq. (7), the signs of τ_1/τ_0 and k_1/k_0 should be the same where anisotropy is large.

Expressions for the electrical conductivity and ordinary Hall effect in cubic metals have been derived by expanding k, τ, and $dk/d\mathcal{E}$ in Kubic harmonics.[14,15] The results of Cooper and Raimes show that when τ_1/τ_0 and k_1/k_0 are of the same sign, the magnitude of R_0 is an increasing function of the magnitude of the anisotropy τ_1/τ_0. In iron and 4% Ni in iron the thermal scattering is dominant and the positive values of R_0 at all temperatures indicate hole conduction is dominant. The hole pockets show a large deviation from spherical surfaces, judging by the large magnitudes of k_1/k_0. Thus R_0 increasing with temperature and then possibly decreasing would be expected, if the anisotropy effects were more important than the effect of the relative sizes of hole and electron pockets. The data show an increase and then a leveling off or a slow decrease near room temperature. It should be noted, however, that the results of Cooper and Raimes should be applied with care whenever deviations from spherical symmetry become fairly large.

The samples containing 15 and 20% Ni show a relatively slow temperature variation of R_0 when expressed in terms of n^*. The large negative values are, as indicated above, fairly close to what is expected from the $6\Gamma_+$ pocket with a small hole contribution. Since this $6\Gamma_+$ pocket is highly spherical and the impurity content is large, only a small anisotropy of the relaxation time is to be expected. And since the impurity scattering is large a smaller temperature dependence is reasonable. The temperature dependence that does exist can be attributed to the hole contribution or to the contribution by the less spherical $4\Gamma_-$ electron pockets.

Comparison between the ordinary Hall coefficients of Fe-Ni measured here and those measured in Fe-Co alloys[5] of corresponding electron number show that the two systems have roughly the same composition dependence when plotted against electron number. There is a difference, however, in that negative, or electron-like, values are reached at lower electron numbers in the Fe-Co system and that the temperature dependence is also less marked there.

ELECTRICAL RESISTIVITY

The measured electrical resistivity is shown as a function of temperature and Ni content in Fig. 3. The data have no unusual aspects, except perhaps for the fact that the rate of change of the resistivity with Ni content becomes much smaller at about 15% Ni. There is, in fact, actually a decrease in resistivity at all temperatures between 15 and 20% Ni. In this range of concentration there should be no ordering and a monotonic increase in resistivity with Ni content would be expected. However, the phase diagrams and the behavior of the samples in preparation indicate that mixed phases are likely by 15% and almost certainly by 20% Ni. According to the magnetic measurements of Hoselitz[18] in an alloy of 84% Fe and 16% Ni there is a tendency for segregation, with the Ni content being larger than the average in the fcc regions and smaller than the average in the bcc regions. A decrease in the resistivity could then occur if the increase in the resistivity in the fcc regions is more than offset by the decrease in the bcc regions, the net result being measured as a decrease in resistivity as phase mixing sets in.

EXTRAORDINARY EFFECT

The spontaneous Hall coefficient R_s was given its first somewhat successful theoretical treatment by Karplus and Luttinger.[19] Their treatment led to the prediction that R_s should vary as the square of the electrical resistivity ρ. This was more or less verified in several cases, but did not seem to be the complete picture, particularly at low resistivities. Subsequently, Smit,[20] Luttinger,[21] and others[22–26] have proposed extensions, corrections, or alternate treatments and have shown that terms other than a ρ^2 dependence should be included. However, the magnitudes of the various terms cannot be calculated theoretically, and it is still not clear which, if any, of the various modifications adequately describes the true situation.

The experimental values of the spontaneous Hall coefficients R_s are shown plotted against the resistivity

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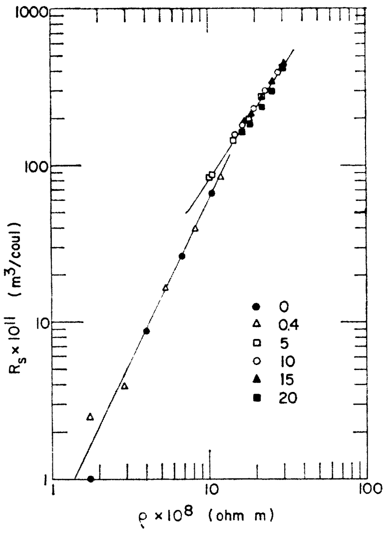

FIG. 5. The spontaneous Hall coefficient as a function of resistivity for iron-nickel alloys, plotted on a log-log scale. The key gives the atomic percent nickel in iron.

on a log-log scale in Figs. 5 and 6, and as a function of temperature in Fig. 2. The log-log presentation is used to test the applicability of the Karplus-Luttinger treatment[19] to this system. The points corresponding to samples with appreciable impurities (5, 10, 15, and 20% Ni) fall more or less on the same straight line of Fig. 5, whereas for the pure and nearly pure (0.4% Ni) samples the data points lie on or near another straight line. If a law of the form ρ^n is assumed for this data, then the values of n are about 1.46 for the impure samples and 2.05 for the "pure" samples.

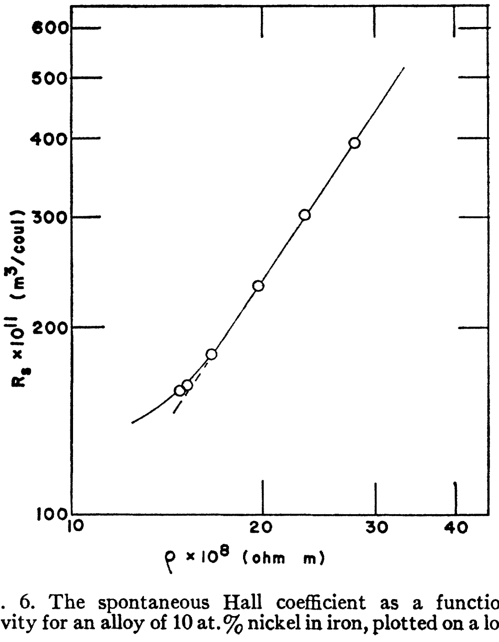

FIG. 6. The spontaneous Hall coefficient as a function of resistivity for an alloy of 10 at.% nickel in iron, plotted on a log-log scale.

Figure 6 is an expanded plot of the data for the sample containing 10% Ni, which in its behavior is representative of the other impure samples as well. The figure shows that the data follow a straight line on a log-log plot, except at the lowest temperatures where the slope decreases. This behavior seems to be typical of a number of alloys, and the general rule seems to be that the greater the relative proportion of impurity to thermal scattering, the greater is the deviation of the slope from 2, this deviation being in the direction of smaller slope. This is in at least qualitative agreement with the model of Luttinger,[21] in which the contribution of the impurity scattering to R_s is proportional to the density of impurities, or to the impurity resistivity in dilute alloys, if Matthiessen's rule holds.

To see if both kinds of contribution might occur, the data were fitted using the method of least squares by an expression of the form

$$R_s = a_1 + b_1\rho_i + c_1\rho^2, \qquad (10)$$

where a_1, b_1, and c_1, are constants, ρ is the total resistivity, and ρ_i the impurity scattering contribution when the resistivity data are fitted by Matthiessen's rule. The constants in (10), as well as in (11) and (12), utilized the whole range of composition measured and all temperatures except 87°K and room temperatures. A term like a_1 arises in the Karplus-Luttinger theory as a consequence of the deviation of the effective internal field from B. The results of this attempt gave a rather good fit to the data even at room temperatures (which is an extrapolation) except at the lowest resistivities.

A second fit to the data was attempted by making the constant term of Eq. (10) zero; that is,

$$R_s = b_2\rho_i + c_2\rho^2. \qquad (11)$$

This resulted in a considerable improvement at the lower resistivities at the expense of a slightly larger average deviation at the higher resistivities. On the whole, Eq. (11) seems to represent the data as well as Eq. (10) does. This is consistent with the de Haas-van Alphen measurements of Anderson and Gold[27] and the theoretical analysis of Kittel[28] in which the effective field is shown to be B.

A third model was also considered in an attempt to fit the data. This model was based partly upon the work of Smit[27] in which the thermal scattering contributes as the square of the thermal scattering term ρ_t of the resistivity. For this reason we have tried an expression of the form

$$R_s = a_3\rho_t^2 + b_3\rho_i + c_3\rho^2. \qquad (12)$$

The results of this attempt were somewhat different from those of Eq. (11), but no significant over-all improvement was apparent.

From the fairly good success in fitting the data over

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a wide range of both impurity and thermal scattering in various proportions and magnitudes, it appears that an expression of the form ρ^n, where $n\neq 2$, is not too significant, since it breaks down over a wide range of data. The slope of 1.4 in Fig. 5 may be due to a mixture of Luttinger and Karplus-Luttinger–type terms.

An interesting correlation occurs between the quantity R_s/ρ^2 at room temperature over the whole Fe-Ni composition range, as reported by Jellinghaus and de Andres,[29] and the first magnetic anisotropy constant K_1.[30] Both are positive from iron to about 80% Ni where both change sign, and their behaviors are very similar over the whole range of compositions. This may be due to the fact that, in the Karplus-Luttinger treatment, which probably gives the dominant contribution at room temperature, R_s/ρ^2 should be proportional to the strength of the spin-orbit coupling and the anisotropy energy should have the same dependence.

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Propagation of Bragg-Reflected Neutrons in Large Mosaic Crystals and the Efficiency of Monochromators

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The general integral equations for describing the multiple Bragg reflections in a mosaic crystal are given. The solutions of the general problem are in the form of infinite series. For the particular problem of a delta-function beam striking a semi-infinite crystal, these series are summed to give modified Bessel functions as the appropriate solutions. The modified Bessel-function solution serves as a Green's function for the generation of solutions of a general beam striking a semi-infinite crystal. Discussed in some detail are the effects of absorption and of crystal-cutting angle on the efficiency of monochromating crystals. The concept of the asymptotic value of neutron current density is stressed to show the importance of not collimating the beam between the neutron source and the monochromating crystal.

I. INTRODUCTION

The approach taken by this paper in analyzing the multiple-Bragg-scattering problem in mosaic crystals is similar to that taken by Hamilton.[1] However, the differential equations given by Hamilton describing the conservation of neutrons are recast in integral form. Consequently, the starting point of the analysis here closely parallels that given by Vineyard.[2] While the historic interest in the multiple-Bragg-scattering problem (or secondary-extinction problem) has been the need to make corrections for it in comparing intensities in determining crystallographic structure, our primary interest has been in the design of diffraction experiments to make greater use of the neutron source available at low- to medium-flux reactors.

For example, in order to obtain an acceptable resolution in doing single-crystal experiments the divergence of the "monoenergetic" beam from a monochromator must be less than of the order of 1° in the horizontal plane and perhaps 3° in the vertical plane (owing to crystal size). This implies that only those neutrons incident on the monochromator in a small solid angle of the order of 0.0006 sr are useful. This solid angle subtends only about 15 sq in. at a reactor face 12 ft away. Thus, for these small angles the reactor face looks like a plane source, and consequently, the inverse-square law does not apply. If it were possible to reflect all the neutrons in this small solid angle $\Delta\Omega$ having energies in the small spread ΔE immediately at the surface of the monochromator, we would have a plane source of "monoenergetic" neutrons which could then be colli-

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