Galvanomagnetic Effects, Magnetostriction, and Spin-Orbit Interaction in Cu-Ni-Fe and Other Ferromagnetic Nickel Alloys*

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In the triangular diagram of Cu-Ni-Fe alloys, the line of zero linear magnetostriction λ_s approximately coincides with the line of zero extraordinary Hall constant R_s and may be interpreted as the line where a $3d$ orbital degeneracy crosses the Fermi level. The "ferromagnetic anisotropy of resistivity" $(\rho_{11}-\rho_1)/\rho_0$ of quenched Cu-Ni-Fe alloys containing more than 60 wt% Ni has been measured at 20°K, where impurity scattering dominates. The large anisotropy values observed in Ni-Fe alloys decrease gradually by addition of copper. In particular, the anisotropy decrease along the line $\lambda_s=0$ suggests that the orbital degeneracy is partially lifted by the addition of copper. This line differs from a line of constant electron concentration, indicating a departure from the rigid-band model. A $3d$ band model with separate iron, nickel, and copper sub-bands correctly predicts the location of the line if the degeneracy is assumed to be located at the top of the nickel sub-band (bottom of the iron sub-band). The same assumption applies successfully to Me-Ni-Fe alloys, where Me is any metal (Cr, W, Mo, V, etc.) forming a nonmagnetic sub-band above the Fermi level. Existing data show that the extraordinary Hall conductivity $\gamma_{Hs}=R_sM_s/\rho^2$ is roughly proportional to the magnetostriction λ_s for all fcc Ni-Fe, Cu-Ni, and Cu-Ni-Fe alloys at $T\ll T_c$, with a slope $\approx 2.0\times 10^{+9}$ mks.

I. INTRODUCTION

THE electrical resistivity of a ferromagnetic polycrystal depends[1] on the angle between current density **j** and magnetization **M**. The "ferromagnetic anisotropy of resistivity" $\Delta\rho/\rho_0$ is defined by

$$\Delta\rho/\rho_0=(\rho_{11}-\rho_1)/(\tfrac{1}{3}\rho_{11}+\tfrac{2}{3}\rho_1). \quad (1)$$

This anisotropy is caused by spin-orbit interaction. It is given roughly by

$$\Delta\rho/\rho_0\approx(A_{so}/\Delta E)^2+\cdots, \quad (2)$$

where ΔE is an energy difference between branches of the $3d$ band in a vicinity of the Fermi level, and where the dots indicate higher-order terms in the spin-orbit parameter A_{so}. This anisotropy has been measured at 20°K in Fe-Ni, Co-Ni, Cu-Ni, Cr-Ni, and V-Ni alloys by Smit[2] and by Van Elst.[3] It reaches 20% in certain Fe-Ni and Co-Ni alloys having an electron concentration of 27.7 electrons per atom. This large anisotropy value has been explained[4] on the basis of Eq. (2) by assuming that a near degeneracy ($\Delta E\approx A_{so}=0.1$ eV) is present in the $3d$ band structure and happens to cross the Fermi level for that value of the electron concentration. A correspondingly large anisotropy of the thermal resistivity has been observed[5] in a 15% Fe–85% Ni alloy at 4.2°K. (All alloy compositions are given in weight percent.) The magnetoresistance of a 15% Fe–85% Ni single crystal has also been investigated[6] at 20°K. When rotating **M** in the plane normal to **j**, the curve of resistivity versus angle is found to have a considerable content of higher harmonics, again a clear indication[6] of an orbital degeneracy at the Fermi level. As shown elsewhere,[4] the same assumption of a near degeneracy also explains why the linear magnetostriction λ_s and the extraordinary Hall constant R_s both vanish and change sign at 27.7 e/at. in the Fe-Ni and Co-Ni series.

The purpose of the present work is to extend the $\Delta\rho/\rho_0$ measurements to the Cu-Ni-Fe system and to discuss relevant information on λ_s and R_s. Like the Ni-Fe alloys (Permalloy), the Cu-Ni-Fe alloys have important technological applications (Mumetal) due in part to their low magnetostriction. The line $\lambda_s=0$ in the triangular diagram of these alloys is shown[7] on Fig. 1. The line $R_s=0$ may be determined roughly from existing Hall data for Fe-Ni,[8] Cu-Ni,[9] and Cu-Ni-Fe[10] alloys and is seen[11] to run close and parallel to the line $\lambda_s=0$ (Fig. 1). This fact confirms that the relation mentioned above between magnetostriction and extraordi-

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[1] J. P. Jan, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1957), Vol. V, p. 1.

[2] J. Smit, Physica **17**, 612 (1951).

[3] H. C. Van Elst, Physica **25**, 708 (1959). The anisotropy value 14.5% for 70 Ni–30 Fe does not interpolate smoothly with values for other alloys studied by Van Elst or by us and seems to be in error.

[4] L. Berger, Physica **30**, 1141 (1964); Phys. Rev. **138**, A1083 (1965).

[5] L. Berger and D. Rivier, Helv. Phys. Acta **35**, 712 (1962).

[6] L. Berger and S. A. Friedberg, Phys. Rev. **165**, 670 (1968). The magnetic anisotropy torque of very pure nickel has recently been found to have discontinuous derivatives as a function of angle probably for the same reason [J. J. M. Franse, Physica **39**, 477 (1968)].

[7] O. Von Auwers and H. Neumann, Wiss. Veroeffentl. a. d. Siemens-Werken **14**, No. 2, 93 (1935). These authors found the line to be the same for quenched and for annealed alloys. The same holds for other ternary alloys. Note also that the lines $\lambda_{100}=0$ and $\lambda_{111}=0$ for most single-crystal alloys are quite close together.

[8] W. Jellinghaus and M. P. De Andres, Ann. Physik **5**, 187 (1960).

[9] P. Cohen, Ph.D. thesis, Carnegie Institute of Technology, 1955 (unpublished). We use 14°K data only, since the Curie point of these alloys is low.

[10] E. R. Sanford, A. C. Ehrlich, and E. M. Pugh, Phys. Rev. **123**, 1947 (1961); A. C. Ehrlich, J. A. Dreesen, and E. M. Pugh, *ibid.* **133**, A407 (1964). Samples were cooled slowly.

[11] The small discrepancy between the two lines is magnified because we show only part of the triangular diagram.

nary Hall effect in the Fe-Ni and Co-Ni series was more than a coincidence. The line $\lambda_s \approx R_s \approx 0$ then appears as the line where the orbitally degenerate state mentioned above crosses the Fermi level of Cu-Ni-Fe alloys. $\Delta\rho/\rho_0$ measurements in Cu-Ni-Fe may give additional information on the degenerate state.

II. SAMPLE PREPARATION

Samples $A1$–$A11$ (see Table I) were cut from Hall samples used in earlier work.[10] Samples $S1$, $O1$, $B2$, $B3$, and $B4$ were vacuum-melted in an induction furnace from Johnson-Matthey iron and nickel and from Fisher Certified Reagent 99.99% copper. After machining, all samples were homogenized for 24 h at 1200°C under a pressure of 1.2 atm of charcoal-purified helium and cooled to room temperature in the furnace. Then they were heated again for 1 h at 700 or 800°C under helium and quenched in water.

Recent Fe^{57} Mössbauer-effect measurements by Nagarajan and Flinn[12] have shown on a microscopic scale that the precipitation of a copper-rich phase[13] requires at least 10 sec at 600°C to affect 6% of the volume of a 60% Cu–28% Ni–12% Fe alloy. The precipitation process should be much more sluggish[13] in our samples, since they contain more nickel. The cooling in less than 2 sec achieved by water quenching should therefore be quite effective in preventing precipitation even at the atomic level.

Polished cross sections of samples $A5$, $A8$, and $A9$ were scanned with an electron microprobe (spot size, $1\text{–}2\times10^{-3}$ mm) at the Mellon Institute in Pittsburgh. The ratio of nickel concentration to copper concentration was found to fluctuate with a relative mean deviation of, respectively, ±6, ±7, and ±8% of its average value over the length of a given sample. This is considered satisfactory by us.

The copper, nickel, and iron concentrations have been determined (Table I) through chemical analysis by Kissell of the Carnegie-Mellon University Metals Research Laboratory and by Ledoux and Co., New York.

The samples, about $30\times8\times0.8$ mm in size, have their ends pressed laterally against copper strips supplying a dc current of about 5–8 A. Two spring-loaded phosphor-bronze potential probes are located about 10 mm apart, along the flat side of the sample.

The sample and sample holder may be turned 90° around a vertical axis with respect to the horizontal magnetic field, thus making **j** either parallel or normal to **M**. The 3–4-kG field, always parallel to the flat side of the sample, is therefore more than sufficient to achieve saturation. The resistivities $\rho_{\parallel}$ and $\rho_{\perp}$ are measured to ±0.1% with a $K3$ potentiometer and a Leeds and Northrup null detector, while potentiometer and sample currents are inverted simultaneously every few seconds to eliminate parasitic thermoelectric voltages.

[12] A. Nagarajan and P. Flinn, Appl. Phys. Letters **11**, 120 (1967). A. Nagarajan, Ph.D. thesis, Carnegie Institute of Technology, 1967 (unpublished); and (private communication).

[13] W. Köster and W. Dannöhl, Z. Metallkunde **27**, 220 (1935).

TABLE I. $\Delta\rho/\rho_0$ values at 20°K for Cu-Ni-Fe.

Sample	wt% Cu	wt% Ni	wt% Fe	$\Delta\rho/\rho_0$ (%)
$A1$	2.6	95.6	1.5	8.3
$A2$	5.6	91.0	2.8	9.8
$A3$	11.7	71.2	17.5	10.3
$A4$	10.5	83.3	4.9	8.24
$A5$	21.3	69.3	10.0	5.3
$A6$	22.4	71.2	6.2	5.55
$A7$	27.4	69.8	3.3	4.8
$A8$	15.8	70.5	14.7	7.6
$A9$	19.3	69.5	11.6	5.9
$A11$	30.8	61.6	8.4	2.8
$S1$	2.31	82.28	15.37	17.45
$O1$	5.76	74.0	20.13	10.3
$B2$	9.28	81.41	9.91	10.9
$B3$	13.62	76.65	9.88	8.45
$B4$	7.11	67.11	25.89	7.2

III. EXPERIMENTAL RESULTS

The data (Table I) have been obtained at 20°K, since $\Delta\rho/\rho_0$ is found[2] to be considerably larger for impurity scattering than for phonon or magnon scattering. The results are displayed on Fig. 1, and tentative contour lines are drawn for $\Delta\rho/\rho_0$. Coordinates correspond to mass concentrations, though the use of atomic concentrations would little affect the location of the points. Included in Fig. 1 are the $\Delta\rho/\rho_0$ data by Van Elst[3] for Ni-Fe and Cu-Ni alloys and for 4.5%Cu–92.2% Ni–2.4% Fe and 10.8% Cu–84.5% Ni–4.7% Fe.[14]

The alloys of maximum $\Delta\rho/\rho_0$ are only found in a closed region around the alloy 85% Ni–15% Fe. One can speak of a "ridge" starting at this binary alloy and following roughly the line $\lambda_s \approx R_s \approx 0$ of the orbital degeneracy; however, the $\Delta\rho/\rho_0$ value decreases gradually when moving along the ridge in the direction of increasing copper concentration. Thus, it seems that copper atoms tend to smear and to lift partially the near degeneracy. If we used only the first term of Eq. (2), an oversimplification, the decrease of $\Delta\rho/\rho_0$ from the 20 to the 5% contour line (Fig. 1) would correspond to a doubling of the gap ΔE. Apparently, ΔE is still sufficiently small to force the $\lambda_s=0$ line to coincide with the $R_s=0$ line in the whole Cu-Ni-Fe system (Fig. 1).

IV. LOCATION OF DEGENERACY

In the binary Ni-Fe and Ni-Co systems, the alloys corresponding to $R_s \approx \lambda_s \approx 0$ (and to maximum $\Delta\rho/\rho_0$) had[4] roughly the same electron concentration 27.7 e/at., in seeming agreement with the rigid-band model. However, in the ternary Cu-Ni-Fe system, the line $R_s \approx \lambda_s \approx 0$ differs from a line of constant electron concentration (Fig. 1). Actually, when copper is added to Ni-Fe, the alloys with $R_s \approx \lambda_s \approx 0$ are found to correspond

[14] A. C. Ehrlich, J. A. Dreesen, and E. M. Pugh (Ref. 10) have measured the magnetoresistance of several Cu-Ni-Fe alloys, but only in transverse fields.

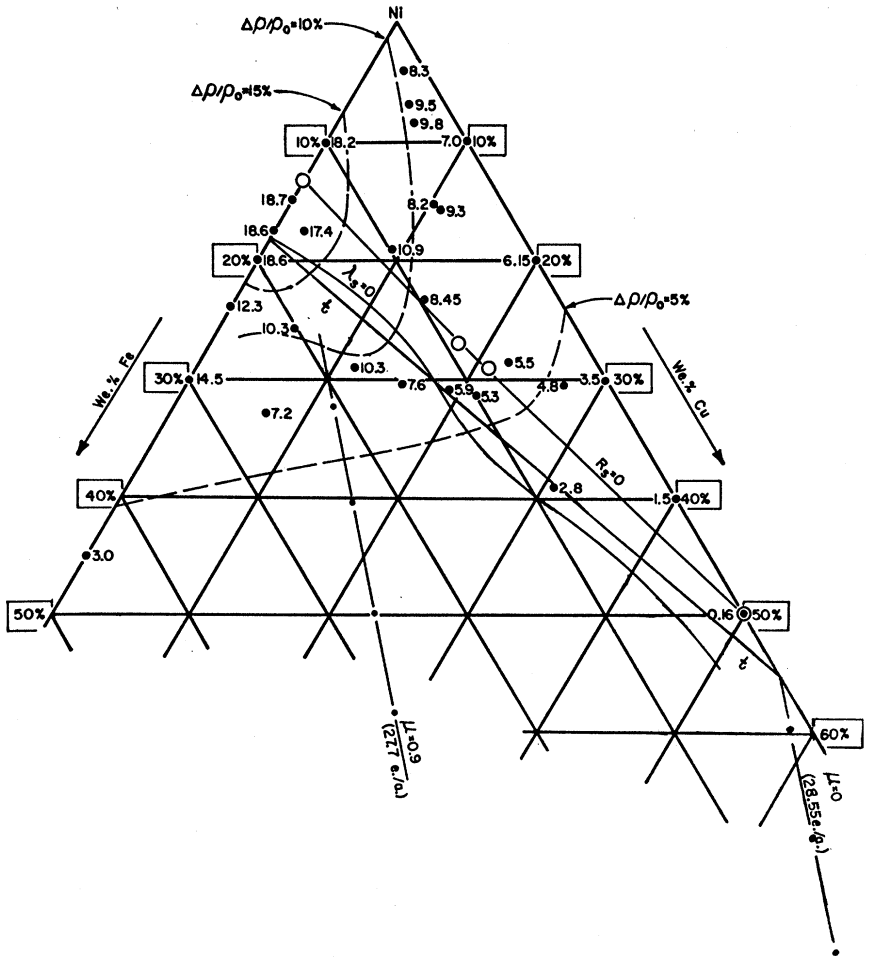

FIG. 1. Triangular diagram for Cu-Ni-Fe alloys. The solid circles and associated numbers indicate measured values of the ferromagnetic anisotropy of resistivity at 20°K, expressed in percent (see also Table I). Dashed lines are tentative contour lines for the anisotropy of resistivity. The open circles indicate four points where the extraordinary Hall coefficient R_s is found by interpolation to vanish and change sign. They have been joined by the solid line $R_s=0$. Another solid curve indicates where the linear magnetostriction λ_s vanishes and changes sign. The solid straight line t is the locus of alloys which have the top T of the nickel sub-band located at the Fermi level. The straight lines with dashes and dots correspond to constant electron concentration and constant number of Bohr magnetons μ.

to larger and larger electron concentration. Note also that the value of $\Delta\rho/\rho_0$ is not a function of electron concentration only.

These facts can be explained on the basis of recent x-ray absorption, photoemission, and reflectivity measurements[15] on Ni-Cu alloys. These measurements show that most features of the $3d$ band remain more or less at a fixed energy for increasing copper concentration, but that the alloy density of states increases at the band bottom and decreases at the top. A similar effect is also predicted by various theorists.[16] This effect should be even larger[16] for Cu-Ni-Fe alloys because of the larger difference of nuclear charge between copper and iron. We picture in Fig. 2(a) a somewhat extreme model, where each alloy component has a separate sub-band. The number of states in the $3d\uparrow$ iron sub-band is equal to five times the atomic concentration C_{Fe} of iron atoms, and a similar relation holds for other sub-bands. Thus, any given feature of the band, such as an orbital degeneracy, will cross the Fermi level at a larger and larger electron concentration value when copper is added.

For example, with this model we can determine when the top T of the $3d\uparrow$ nickel sub-band [Fig. 2(a)] is crossing the Fermi level. The total number of holes in the alloy $3d\uparrow$ band is $0.55+2C_{\mathrm{Fe}}-C_{\mathrm{Cu}}$, where 0.55 is the $4s$ electron concentration. Thus,

$$5C_{\mathrm{Fe}}=0.55+2C_{\mathrm{Fe}}-C_{\mathrm{Cu}}$$

or

$$3C_{\mathrm{Fe}}+C_{\mathrm{Cu}}=0.55. \tag{3}$$

Equation (3) is represented by the straight line t on Fig. 1. This line coincides approximately with the line $R_s\approx\lambda_s\approx 0$, indicating that the orbital degeneracy is located very close to the top of the nickel sub-band of Cu-Ni-Fe alloys. Note that this applies in particular to Ni-Fe alloys, and also to Cu-Ni alloys where T becomes the top of the whole $3d$ band. Note also that iron added to a Ni-Fe alloy tends to raise the Fermi level above T,

[15] D. H. Seib and W. E. Spicer, Phys. Rev. Letters **20**, 1441 (1968); L. V. Azaroff, Science **151**, 785 (1966).

[16] J. Friedel, Nuovo Cimento Suppl. **7**, 287 (1958); J. L. Beeby, Phys. Rev. **135**, A130 (1964); S. Kirkpatrick, B. Velicky, N. D. Lang, and H. Ehrenreich, J. Appl. Phys. **40**, 1283 (1969). Even a small impurity potential will split triplet Fe states off the rectangular top of the Ni band. See *Theory of Magnetism in Transition Metals*, edited by W. Marshall (Academic Press Inc., New York, 1967), p. 309.

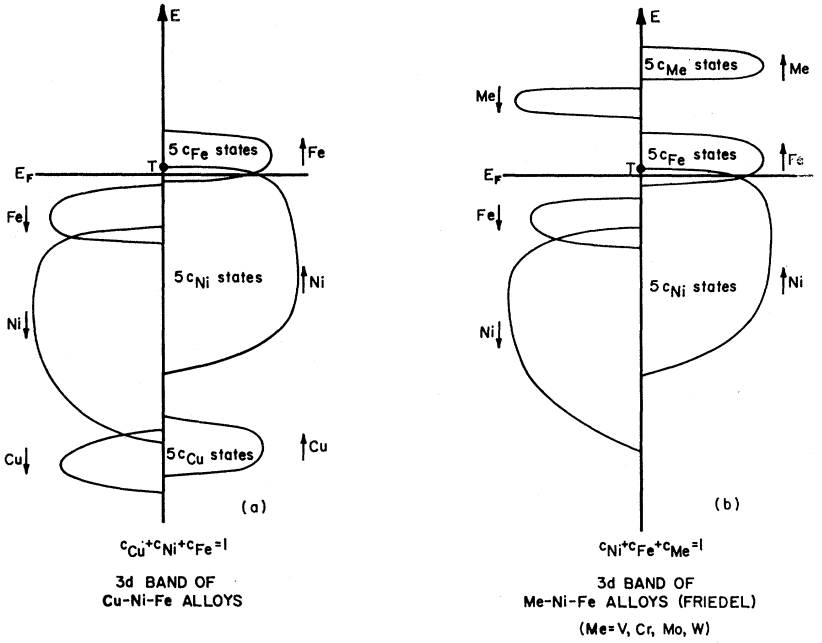

FIG. 2. (a) Model with separate sub-bands for Cu-Ni-Fe alloys. T indicates the top of the $3d\uparrow$ nickel sub-band. (b) Model for Me-Ni-Fe alloys, where Me forms nonmagnetic virtually bound states above the Fermi level, according to Friedel.

and not to depress it, due to the rapid decrease in the number of states contained in the nickel sub-band.

Equation (3) is precisely valid only if the nickel and iron sub-bands do not overlap. Since our theory involves only the boundary T between nickel and iron sub-bands, it is not really necessary for the copper and nickel sub-bands to be distinct, as pictured in Fig. 2(a). Neither should it be assumed that the separation of sub-bands on the energy scale implies a complete spatial localization of the wave function on the corresponding type of atoms. Note also that our statement about the presence of T at the Fermi level for alloys with $\lambda_s \approx R_s \approx 0$ is actually independent of the identification of T with an orbital degeneracy.

If the nickel and iron sub-bands are partially distinct [Fig. 2(a)], their common boundary T may be expected to correspond to a local minimum of the density of states. Even though the sub-bands do not remain rigid when adding iron to pure nickel, the experimental curve of electronic specific heat is found to go down and to exhibit[17] a strong upward concavity in the region around 15% Fe–85% Ni and to have a minimum at 30% Fe. Thus, there is a better experimental verification for the idea of a minimum than for the opposite assumption of a density-of-states maximum introduced by Smit[2] to explain the $\Delta\rho/\rho_0$ maximum.

The present theory may easily be extended to Me-Ni-Fe alloys [Fig. 2(b)], where Me stands for any metal impurity (V, Mo, W, Cr, etc.) forming nonmagnetic $3d$ bands[16,18] (virtually bound states) above the Fermi level, according to Friedel. The band model of Fig. 2(b) is consistent with many saturation-magnetization and neutron-diffraction data for these alloys. A degeneracy located at the top T of the Ni sub-band is predicted to cross the Fermi level when

$$10C_{\mathrm{Me}}+5C_{\mathrm{Fe}}=0.55+2C_{\mathrm{Fe}}-Z\,C_{\mathrm{Me}}$$

or

$$3C_{\mathrm{Fe}}+(10+Z)C_{\mathrm{Me}}=0.55\,, \tag{4}$$

where Z is the valence difference[16] between Me and Ni (-5 for V, -4 for Mo, W, and Cr). Note that Eqs. (3) and (4) coincide if we set formally $Z=-9$ for Cu.

Our Eq. (4) agrees approximately with the general empirical rules formulated by Enoch and Fudge and by Rassmann and Hofmann[19] for the location of the $\lambda_s=0$ line of these Me-Ni-Fe alloys and explains these rules in terms of a definite band model. Some caution is necessary since usually only a portion of the line has been determined, and since the line is probably not quite straight but rather concave towards pure nickel.

[17] W. H. Keesom and B. Kurrelmeyer, Physica **7**, 1003 (1940); K. P. Gupta, C. H. Cheng, and P. A. Beck, J. Phys. Chem. Solids **25**, 73 (1964). A similar decrease is observed when both copper and iron are added [R. Ehrat, A. C. Ehrlich, and D. Rivier, *ibid.* **29**, 799 (1968)]. Complications caused by spin fluctuations are present in Cu-Ni at the critical concentration of 60% Cu. As expected, the decrease is smaller for Co-Ni than for Fe-Ni, because of smaller nuclear charge difference [J. C. Ho and R. Viswanathan, Phys. Rev. **172**, 705 (1968)].

[18] The essential difference between Fe and Me sub-bands is that the former are magnetized by the strong iron intra-atomic exchange.

[19] R. D. Enoch and A. D. Fudge, Brit. J. Appl. Phys. **17**, 623 (1966); G. Rassmann and U. Hofmann, J. Appl. Phys. **39**, 603 (1968); magnetostriction data for various Me-Ni-Fe have been obtained by U. Hofmann, Z. Angew. Phys. **21**, 425 (1966); I. M. Puzej, Izv. Akad. Nauk SSSR Ser. Fiz., **16**, 549 (1952); I. M. Puzej and B. V. Molotilov, *ibid.* **22**, 1244 (1958); W. A. Dean, Rensselaer Polytechn. Inst. Eng. Sci. Ser. **26**, 31 (1929); R. C. Hall, J. Appl. Phys. **30**, 816 (1959). Our theory even extends in a straightforward manner to the quaternary alloys Cu-Ni-Fe-Mo, investigated by F. Pfeifer [Z. Metallkunde **57**, 295 (1966)] and by Enoch and Fudge (Ref. 19).

Corresponding data for the location of the $R_s=0$ line are entirely missing. For binary Me-Ni, Eq. (4) reduces to $C_{Me}=0.55/(10+Z)$, corresponding[16] to alloys of just vanishing atomic moment. This prediction agrees with the experiments of Went,[20] who found λ_s and the atomic moment μ to vanish almost simultaneously in these alloys at 0°K, and is to be expected [Fig. 2(b)], since in the absence of iron the top of the nickel subband is also the top of the whole $3d\uparrow$ band (excluding the nonmagnetic Me states). Similarly, and for the same reason, the line $\lambda_s\approx R_s\approx 0$ in the Cu-Ni-Fe system is seen (Fig. 1) to cross the line of vanishing atomic moment μ at a point located on the Cu-Ni line.

If we accept the conclusion that the degeneracy is located approximately at the top T of the $3d\uparrow$ nickel sub-band of the alloy, we can speculate that it may be related to the Δ_5 degenerate states present at the top of the $3d$ band of pure nickel.[21]

V. GENERAL RELATION BETWEEN EXTRAORDINARY HALL EFFECT AND MAGNETOSTRICTION

One may wonder whether the coincidence of the $\lambda_s=0$ and $R_s=0$ lines mentioned above does not suggest the existence of a more general empirical relation between the Hall effect and magnetostriction. Since existing theories and experiments show[22,4] roughly $R_s\propto\rho^2$ in iron and nickel alloys of sufficiently high resistivity ρ, the extraordinary Hall conductivity $\gamma_{Hs}=R_sM_s/\rho^2$, where M_s is the saturation magnetization, is more meaningful than R_s. We plot γ_{Hs} versus λ_s on Fig. 3, using existing data[7–10,20,23,24] for Ni–Fe, Cu–Ni, and Cu-Ni-Fe. It is likely that a relation between γ_{Hs} and λ_s holds only for $T\ll T_c$. However, the relation $R_s\propto\rho^2$ may break down in dilute alloys at low temperature if ρ is too low. Thus, we use room-temperature data in Fig. 3, with the exception of Cu-Ni alloys where we use 0 or 14°K values because of the low Curie point T_c. The two Cu-Ni-Fe curves correspond to different paths[10] in the triangular diagram of these alloys.

We see that γ_{Hs} is roughly proportional to λ_s for all fcc Cu-Ni, Ni-Fe,[25] and Cu-Ni-Fe for which data exist, with a slope $\approx 2\times 10^9$ mks. The degree of accuracy of the relation is as good as can be expected, given that λ_s and R_s (or even M_s or ρ) were measured on different samples by different investigators.

Such a relation may be expected from the theory if we assume for simplicity that spin-orbit interaction H_{so} mixes the five $3d$ states by pairs (φ_a,φ_b) only. Writing

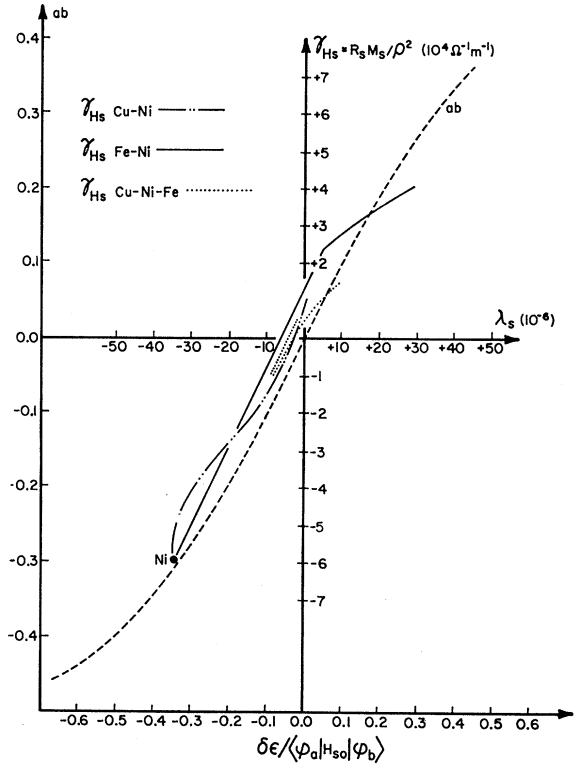

FIG. 3. Dashed curve: theoretical relation between product ab and spin-orbit energy shift $\delta\epsilon$. All other curves: experimental relation between extraordinary Hall conductivity γ_{Hs} and linear magnetostriction λ_s for fcc Ni-Fe, Cu-Ni, and Cu-Ni-Fe alloys. Scales for $\delta\epsilon$ and ab are arbitrary, except that they are chosen to result in equal initial slope for the theoretical and experimental curves.

$\varphi=a\varphi_a+ib\varphi_b$ for a state at a given point of the Fermi surface, one can[4,26] prove $\gamma_{Hs}\propto ab$, where a and b are real. On the other hand, in the presence of a near degeneracy causing a local decrease of the density of states one can prove[4] $\lambda_s\propto\delta\epsilon$ with a deformation-potential theory, where $\delta\epsilon=\langle\varphi_a|H_{so}|\varphi_b\rangle\{1-[1-4(ab)^2]^{1/2}\}/2ab$ is the spin-orbit energy shift at the Fermi level (Fig. 3). For $|ab|\leqslant 0.3$, this last expression gives approximately $\delta\epsilon\propto ab$. Thus $\gamma_{Hs}\propto\lambda_s$, unless the degeneracy is so complete that $|ab|$ would be too close to its maximum value 0.5.

VI. CONCLUSIONS

Our measurements show that the large values of the ferromagnetic anisotropy of resistivity observed in Ni-Fe alloys around 15% Fe decrease gradually by addition of copper. This suggests that the orbital degeneracy present in the band of the binary alloys is broadened and partially destroyed by the copper atoms.

By combining existing data on linear magnetostriction λ_s and extraordinary Hall constant R_s, it is shown

[20] J. J. Went, Physica **17**, 98 (1951).
[21] L. Hodges, D. R. Stone, and A. Gold, Phys. Rev. Letters **19**, 655 (1967).
[22] R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954); J. M. Luttinger, *ibid.* **112**, 739 (1958); W. Köster and W. Gmöhling, Z. Metallkunde **52**, 713 (1961); W. Köster and O. Romer, *ibid.* **55**, 805 (1964).
[23] A. Schulze, Z. Physik **50**, 448 (1928).
[24] M. Yamamoto and T. Nakamichi, Sci. Rep. Res. Inst. Tohoku Univ. **A11**, 168 (1959).
[25] A similar relation exists in the fcc Co-Ni system for the same reason. It is less accurate, perhaps because the Co and Ni subbands are not very distinct.
[26] Similarly, $\langle L_z\rangle\propto ab$.

that the lines $\lambda_s=0$ and $R_s=0$ coincide approximately in the triangular diagram of Cu-Ni-Fe alloys and probably represent the line where the degeneracy crosses the Fermi level. By use of a separate-band model of alloys, it is shown that the degeneracy is located close to the top T of the nickel $3d\uparrow$ sub-band (bottom of the iron sub-band). The same model predicts correctly the location of the $\lambda_s=0$ line of Me-Ni-Fe alloys, where Me=V, W, Mo, Cr, etc.

Note that the identification of the degeneracy with T would probably never have been obvious if we had restricted our investigation to the binary alloys.

By use of existing data, it is even shown that the extraordinary Hall conductivity $\gamma_{Hs}=R_sM_s/\rho^2$ is approximately proportional to λ_s for all fcc Ni-Fe, Cu-Ni, and Cu-Ni-Fe alloys at $T\ll T_c$. Both λ_s and γ_{Hs} are due to spin-orbit interaction.

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Calculation of the Generalized Watson Sums with an Application to the Generalized Heisenberg Ferromagnet*

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A method of calculating the generalized Watson sums $\Phi(\alpha,\eta)=\sum_{\mathbf{k}}(e^{\alpha[1-\eta J(\mathbf{k})/J(0)]}-1)^{-1}$ is presented. These sums over all reciprocal-lattice vectors in the first Brillouin zone are evaluated by approximating them with integrals which are then calculated analytically for $0\leq\alpha\leq\infty$ and $0\leq\alpha\leq1$. To demonstrate the applicability of this technique, the magnetization of an anisotropic Heisenberg ferromagnet is calculated for the simple cubic (sc), bcc, and fcc lattices.

I. INTRODUCTION

THE evaluation of the generalized Watson sums[1–5] has been the subject of several papers over the past 30 years. The sums have the form

$$\Phi(\alpha,\eta)=\frac{1}{N}\sum_{\mathbf{k}}\left\{\exp\left[\alpha\left(1-\eta\frac{J(\mathbf{k})}{J(0)}\right)\right]-1\right\}^{-1}. \quad (1)$$

Here $J(\mathbf{k})$ is defined by

$$J(\mathbf{k})=\sum_{\mathbf{k}}J_{\mathbf{g},\mathbf{f}}e^{i\mathbf{k}\cdot(\mathbf{g}-\mathbf{f})}, \quad (2)$$

where $J_{\mathbf{g},\mathbf{f}}$ is an exchange constant corresponding to the sites $\mathbf{g}$ and $\mathbf{f}$ on a given lattice. The sum in Eq. (1) is taken over all reciprocal-lattice vectors in the first Brillouin zone; α and η are parameters. In this paper we will limit the discussion to

$$0\leq\alpha\leq\infty, \quad 0\leq\eta\leq1. \quad (3)$$

The sum (1) arises in the theory of magnetism and other cooperative phenomena.[6] It must be evaluated in order to calculate the thermodynamic quantities of interest as a function of temperature. [As illustrated in Sec. III, the temperature dependence of the sum (1) is contained in parameter α.]

Previous calculations of $\Phi(\alpha,\eta)$ have been mostly based on series expansions, and are therefore valid only over limited temperature regions. The purpose of this paper is to describe a method of evaluating $\Phi(\alpha,\eta)$ without necessitating the use of series expansions or extensive computer summations. It is shown that an analytic expression for $\Phi(\alpha,\eta)$ in terms of α and η can be found, which is valid for all values of α. It is hoped that the methods exhibited here will be found useful for

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[1] G. N. Watson, Quart. J. Math. **10**, 266 (1939).

[2] M. Tikson, J. Res. Natl. Bur. Stds. **50**, 177 (1953).

[3] A. A. Maradudin, E. W. Montroll, G. H. Weiss, R. Herman, and H. W. Milnes, Acad. Roy. Belgique Classe Sci. Mem. **XIV**, 7 (1960).

[4] A. Levitas and M. Lax, Phys. Rev. **110**, 1016 (1958).

[5] I. Mannari and C. Kawabata, Research Notes of Dept. of Physics, Faculty of Science, Okayama University, Okayama, Japan, Report No. 15, 1964 (unpublished).

[6] See for example R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. **127**, 95 (1962).