Effects of Compensation on the Galvanomagnetic Properties of Nonmagnetic and Ferromagnetic Metals

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Measurements of the galvanomagnetic properties of nickel in the high-field region are described. For a general field direction the magnetoresistance saturates and the Hall coefficient is linear with the field and corresponds within experimental error to one electron per atom. This behavior is at variance with the systematic rules for the observed galvanomagnetic properties of nonmagnetic metals, from which one expects nickel to behave like a compensated metal, having a quadratic magnetoresistance and a vanishingly small Hall coefficient at sufficiently high fields. The theoretical explanation of these rules has been known qualitatively for some time, but is here developed rigorously and shown to account for the available experimental data for nonmagnetic metals in the high-field region. The theory is extended to ferromagnetic metals by considering the effect of removing the spin degeneracy of the energy bands. It is shown that the behavior of nickel can then be explained in terms of a simple model in which one spin-up (higher energy) sheet of the Fermi surface in a single spin zone of the d band has an electron character. The observed approach to compensated behavior of iron is consistent with the theory, and indicates that an alternative explanation of the behavior of nickel in terms of a low mobility in the d bands is unlikely to be true.

I. INTRODUCTION

THE relation between the position of a metal in the periodic table and the field dependence of its transverse magnetoresistance measured in high magnetic fields at low temperatures has been qualitatively understood for many years. A simple two-band model predicts that the magnetoresistance will saturate at sufficiently high fields, unless the metal is compensated (i.e., the number of electrons and holes per unit volume is identical). The magnetoresistance will then increase quadratically with the field. The latter behavior has been observed in all even-valent metals (and also in odd-valent metals having an even number of atoms in the primitive unit cell). The compensation of these metals has been attributed correctly to the fact that the capacity of the Brillouin zone is two atoms per unit cell. In crude terms, the number of electrons "spilling over" into zones of high energy must equal the number of holes left unoccupied in zones of lower energy.

The galvanomagnetic properties of nickel reported in a previous publication[1] appear to be at variance with this general behavior. Although nickel is even-valent, the magnetoresistance saturates for a general field direction, while the Hall voltage is linear and corresponds roughly to one electron per atom in fields up to 18 kOe. It was thought that this behavior was due to the low mobility of the holes in the bands derived from the d states of the free atom compared with the mobility of the electrons in the s band, each numbering approximately one per atom. This explanation is now thought to be unlikely, since more recent measurements in applied fields up to 80 kOe give essentially the same results.[2] This shows that an unreasonably low mobility in the d band relative to the s band would be required.

An alternative explanation of the galvanomagnetic properties of nickel is proposed in this paper. The exchange interaction responsible for the ferromagnetism of nickel resolves the degeneracy of the two spin states per unit cell of opposite signs, the capacity of the Brillouin zone for each being unity. We shall show that if one, and one only, of the spin-up (higher energy) zones derived from a d band by the exchange splitting (called a *spin zone* and having a capacity of one electron per unit cell) contains a sheet of the Fermi surface having the topology of an electron surface, then the metal will be uncompensated and will have a Hall coefficient in the high-field region corresponding to exactly one electron per atom.

In Sec. II, we shall derive rigorously the results concerning the compensation of nonmagnetic metals, which have been tacitly understood for some time, and relate them systematically to the observed galvanomagnetic properties of those metals that have been measured in the high-field region. In Sec. III, the theoretical discussion is extended to ferromagnetic metals, and, in Sec. IV, the experimental data for nickel (and preliminary results for iron) are presented and interpreted.

II. COMPENSATION IN NONMAGNETIC METALS

For any Bravais lattice the capacity of the corresponding Brillouin zone for electrons of one sign of spin is one quantum state per primitive unit cell of the lattice. In a nonmagnetic metal the two spin states of opposite sign are degenerate, so the total capacity of each Brillouin zone is two states per unit cell. The number of electrons per unit cell available to occupy these states equals the product of the atomic number Z and the number s of atoms per unit cell.

Let us consider now the distribution of these sZ electrons among the various Brillouin zones. We shall distinguish by subscripts f, i, and j, respectively, between zones which are completely occupied, zones in which the topology of the corresponding sheet of the Fermi sur-

[1] E. Fawcett and W. A. Reed, Phys. Rev. Letters **9**, 336 (1962).
[2] W. A. Reed and E. Fawcett, Bull. Am. Phys. Soc. **8**, 247 (1963).

face has an electron character, and zones in which the sheet has a hole character. There is no ambiguity in determining the character of a multiply connected sheet if one adopts the following operation definition: The sheet has an electron/hole character if, for a general field direction, every closed cyclotron orbit (defined by a closed line of intersection with the sheet of a plane perpendicular to the field) encloses a region of occupied/unoccupied states, or is itself enclosed by such an orbit on the same sheet.[3]

The proviso stipulated in this definition, that the magnetic field be in a general direction, is needed to exclude field directions along the symmetry axes, where, for example, the multiply connected electron sheet of copper[4] supports hole orbits not enclosed by electron orbits. The proviso excludes also field directions lying in symmetry planes, where the same sheet supports periodic open orbits, and field directions in a solid angle around each symmetry axis, where it supports aperiodic open orbits.[5,6] In the rest of the paper the magnetic field is assumed to be in a general direction in this sense, unless otherwise explicitly stated.

The last proviso is needed to include by the definition such surfaces as the hole sheet in the second zone of Mg,[7] which supports electron orbits enclosed by hole orbits when the field is near the hexagonal axis. The fourth zone in the free-electron model of Sn[8] is another interesting example of electron orbits enclosed by hole orbits on the same sheet, which illustrates the fact that an electron and hole sheet cannot exist in the same zone according to this definition. Even though in this case the apparent electron sheet is simply connected, it is completely enclosed by the multiply connected hole sheet.[9]

We can now write down the equation for the total number of occupied states,

$$sZ=\sum_f 2_f+\sum_i n_i^e+\sum_j(2_j-n_j^h)\,, \tag{1}$$

and define the algebraic sum,

$$n_A=-\sum_i n_i^e+\sum_j n_j^h=-n^e+n^h\,, \tag{2}$$

where the total numbers of electrons and holes per unit cell are $n^e=\sum_i n_i^e$ and $n^h=\sum_j n_j^h$, respectively. For any branch of the energy band structure in which the energy is not a monotonic function of the wave vector, it is possible for several nonequivalent sheets of either character (but not both as pointed out above) to exist in the same zone (e.g., the three electron sheets in the third zone of Mg[7]). If these sheets in the same zone have hole characters, the factor 2 in the j summation of Eq. (1) should be counted only once.

The quantity n_A defined in Eq. (2) can be determined experimentally from the Hall coefficient A_H measured in the high-field region,[10] by means of the expression

$$n_A=\Omega/A_Hq\,, \tag{3}$$

Ω being the atomic volume and $-q$ the electron charge. Equation (3) is not valid for field directions which permit open orbits on a multiply-connected sheet of the Fermi surface, nor for $n^e=n^h$, when, in fact, the vanishing of the Hall terms in the conductivity tensor leads to a transverse-even voltage which in the high-field region dominates the Hall voltage, the latter having no direct relation to n_A.[11–13] Equation (3) is not valid also if magnetic breakdown[14] occurs, since with breakdown some cyclotron orbits traverse two or more sheets of the Fermi surface in different zones.

Adding Eqs. (1) and (2), we obtain

$$-n_A=sZ-2(F+J)\,, \tag{4}$$

where F is the number of completely full zones and J is the number of zones containing hole sheets. A metal is said to be compensated when $n^e=n^h$, i.e., $n_A=0$. According to Eq. (4) this can *never* be the case when sZ is *odd*, since n_A must then equal a positive or negative odd integer. On the other hand, if sZ is *even*, n_A can equal a positive or negative even integer, or the metal *can* be compensated if

$$2J=sZ-2F\,. \tag{5}$$

A compensated metal can be characterized experimentally in the high-field region by the transverse-even voltage (e.g., Ga [15] and Zn [16]), or by the quadratic field dependence of the transverse magnetoresistance. Since the latter is a more readily determined characteristic, which contrasts strongly with the saturation of the transverse magnetoresistance of an uncompensated metal, it is used here as a criterion for compensation. Table I summarizes the predicted values of n_A from Eq. (4) and the observed behavior for the metals that have been

[3] E. Fawcett, Phys. Rev. Letters **6**, 534 (1961).
[4] A. B. Pippard, Phil. Trans. Roy. Soc. (London) **A250**, 325 (1957).
[5] I. M. Lifshitz and V. G. Peschanskii, Zh. Eksperim. i Teor. Fiz. **35**, 1251 (1958) [translation: Soviet Phys.—JETP **8**, 875 (1959)].
[6] M. G. Priestley, Phil. Mag. **5**, 111 (1960).
[7] L. M. Falicov, Phil. Trans. Roy. Soc. (London) **A255**, 55 (1962).
[8] A. V. Gold and M. G. Priestley, Phil. Mag. **5**, 1089 (1960).
[9] Illustrations of the Fermi surfaces of Cu, Mg, and Sn can be found, for example, in the review of the de Haas–van Alphen effect in these and other metals by D. Shoenberg in *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 74.
[10] The high-field region in this context means the region of high magnetic fields where $\omega_c\bar{\tau}\gg1$ for all cyclotron orbits, ω_c being the cyclotron frequency and $\bar{\tau}$ the relaxation time averaged over the orbit. This region can normally be achieved only in pure metals at low temperatures.
[11] I. M. Lifshitz, M. Ya. Azbel, and M. I. Kaganov, Zh. Eksperim. i Teor. Fiz. **30**, 220 (1955) [translation: Soviet Phys.—JETP **3**, 143 (1956)].
[12] I. M. Lifshitz, M. Ya. Azbel, and M. I. Kaganov, Zh. Eksperim. i Teor. Fiz. **31**, 63 (1956) [translation: Soviet Phys.—JETP **4**, 41 (1957)].
[13] R. G. Chambers, Proc. Roy. Soc. (London) **A238**, 344 (1956).
[14] M. H. Cohen and L. M. Falicov, Phys. Rev. Letters **7**, 231 (1961).
[15] W. A. Reed and J. A. Marcus, Phys. Rev. **126**, 2398 (1962).
[16] W. A. Reed and G. F. Brennert, Phys. Rev. **130**, 565 (1963).

TABLE I. Compensation in nonmagnetic metals.

Group	Metals	s[a]	Z_2[b]	$sZ-2F_0$[c]	N[d]	J[e]	n_A[f] theory	n_A[g] experiment
IA	Li,Na,K	1	1	1	1	0	−1	−0.89 to 0.96[h]
IB	Cu,Ag	1	1	1	1	0	−1	−0.93 to 0.96[i]
IIB	Mg,Zn,Cd	2	0	4	4	2	0	compensated
III	Al	1	1	3	1	1	+1	+0.93[h]
	Ga	4	1	12	(8	4)	0	compensated
	In	1	1	3	(1	1)	+1	+0.95[h]
	Tl	2	1	6	(4	2)	0	compensated
IV	Sn	2	0	8	(4	2)	0	compensated
	Pb	1	0	4	2	3	0	compensated
V	Bi	2	1	10	(2	1)	0	compensated
VI	Mo,W	1	0	6	(4	2)	0	compensated
VII	Re	2	1	14	(2p	p)	0	compensated
VIII	Pt,Pd	1	0	10	(2p	p)	0	compensated

[a] s is the number of atoms per primitive unit cell.
[b] Z_2 is the atomic number Z modulo 2.
[c] See Eq. (6) for a definition of F_0.
[d] See Eq. (7) for a definition of N; p is an integer.
[e] See Eq. (4) for a definition of J.
[f] The theoretical value of n_A from Eqs. (4) and (7) is $2J-N$.
[g] The compensated metals are determined experimentally by the criterion of a nonsaturating field-dependence of the transverse magnetoresistance, approaching quadratic in the high-field region, for a general field direction: Mg (See Ref. 18), Zn (See Ref. 16), Ga (See Ref. 15), Tl (See Ref. 19), Bi (See Ref. 20), Re (see Ref. 21), Pd (see Ref. 22), and Fawcett (see Refs. 3, 17, and 23) gives references to the other compensated metals.
[h] Chambers and Jones (Ref. 24).
[i] Taylor, Merrill, and Bowers (Ref. 25) also give $n_A=-1.36$ for Au. However, their sample had a low-resistance ratio, $R_{RT}/R_{4.0K}=100$, and Chambers (Ref. 13) found a value much closer to, but still less than, minus one for a sample having a higher resistance ratio 825. Both authors used polycrystalline samples, which may give erroneous results since the electron surfaces for all the Group IB metals are multiply connected and support hole orbits for some field directions.

measured in the high-field region.[15–25] According to this criterion of compensation all the metals for which sZ is even are in fact compensated,[3,17] as indicated in column g.

In column (c) of Table I, $sZ-2F_0$ is given for a value F_0 of F corresponding to the total number of closed shells of the free atom ($F_0=1$ for an s shell, 3 for a p shell, etc.) for the s atoms in the unit cell. For all but the Group VIII metals we find that

$$sZ-2F_0=sV, \tag{6}$$

where V is the maximum chemical valency of the metal. The true value of F will be greater than F_0 if pairs of "valence" electrons completely fill one or more inner-Brillouin zones in the metallic state. This will usually be the case if $sV\gg1$, either due to a large value of s(e.g., Ga) or to a large value of V (Group IV and higher).

The total number of occupied states per unit cell in partially-filled Brillouin zones,

$$N=sZ-2F, \tag{7}$$

cannot, in fact, be determined from galvanomagnetic measurements alone. The values of N and J listed in colums (d) and (e) of Table I are in accordance with our current knowledge of the band structures of these metals determined mainly by other experimental techniques. The bracketed values are still only tentative since the corresponding band structures are not well understood. In such a case J and N are chosen to give a value of n_A equal to zero for a compensated metal, and a positive or negative integer nearest the experimental value given in column (g) of Table I for an uncompensated metal.

We wish to emphasize that for every metal listed in Table I the galvanomagnetic properties are qualitatively consistent with the theoretical ideas implicit in Eqs. (1), (2), and (3). The only quantitative discrepancy is for the uncompensated metals which, when measured with high accuracy by the inductive method,[24,26] yield values of $|n_A|$ systematically lower by a few percent than the expected integral values. This discrepancy may be due to an undetected systematic error in the method. But if this is not the case, Chambers and Jones[24] suggest that the discrepancy may be a genuine indication of the shortcomings, due to electron-electron interactions, of the one-electron model upon which Eq. (1) is based.

III. COMPENSATION IN FERROMAGNETIC METALS. THEORY

The resolution of the spin degeneracy by magnetic interactions will not change the terms in Eq. (1) if the splitting of the energy levels for states of opposite spin is sufficiently small. But in a ferromagnetic metal where the exchange interaction is comparable to the width of an energy band, we clearly must distinguish between the characters of the two sheets of opposite spin derived from each sheet of the Fermi surface of the unperturbed metal. We must also consider the possibility that a Brillouin zone may be completely occupied (or empty) for electrons of one spin, while being partially occupied for electrons of the opposite sign of the spin. To avoid confusion we shall call the assembly of quantum states in a Brillouin zone of one sign of spin, a *spin zone*; the capacity of a spin zone is one electron per unit cell.

When these possibilities are included in the formalism, Eqs. (1) and (2) become

$$sZ=\sum_{f\uparrow}1_f+\sum_{f\downarrow}1_f+\sum_{i\uparrow}n_i^e(\uparrow)+\sum_{i\downarrow}n_i^e(\downarrow)$$
$$+\sum_{i\uparrow}[1_i-n_j^h(\uparrow)]+\sum_{j\downarrow}(1_j-n_j^h(\downarrow)] \tag{8}$$

[17] E. Fawcett, Phys. Rev. Letters **7**, 370 (1961).
[18] R. W. Stark, T. G. Eck, W. L. Gordon, and F. Moazed, Phys. Rev. Letters **8**, 360 (1962).
[19] N. E. Alekseevskii and Y. P. Gaidukov, Zh. Eksperim. i Teor. Fiz. **43**, 2094 (1962) [translation: Soviet Phys.—JETP **16**, 1481 (1963)].
[20] S. Mase and S. Tanuma, J. Phys. Soc. Japan **14**, 644 (1959).
[21] W. A. Reed and E. Fawcett, Bull. Am. Phys. Soc. **7**, 478 (1962).
[22] N. E. Alekseevskii, V. S. Egorov, G. E. Karstens, and B. N. Kazak, Zh. Eksperim. i Teor. Fiz. **43**, 731 (1962) [translation: Soviet Phys.—JETP **16**, 519 (1963)].
[23] E. Fawcett, Phys. Rev. **128**, 154 (1962).
[24] R. G. Chambers and B. K. Jones, Proc. Roy. Soc. (London) **A270**, 417 (1962).
[25] M. T. Taylor, J. R. Merrill, and R. Bowers, Phys. Rev. **129**, 2525 (1963).
[26] F. E. Rose, M. T. Taylor, and R. Bowers, Phys. Rev. **128**, 1122 (1962).

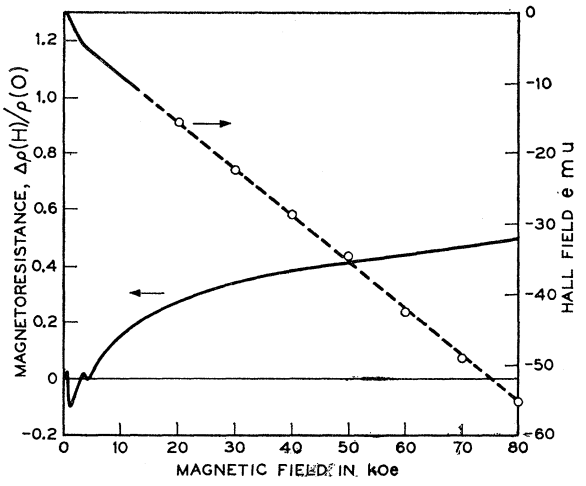

FIG. 1. Magnetoresistance and Hall field of nickel with the magnetic field H in the direction for which there are no open cyclotron orbits. The nickel sample, with its axis along the <110> symmetry axis, has a resistance ratio, $\rho_{295°K}/\rho_{4.2°K}=2700$, in zero applied field. The magnetic field up to 18 kOe was applied at an angle $\varphi=15°$ to the equatorial plane of the sample, and from 18 to 80 kOe at $\varphi=6°$. The measured Hall field has been corrected accordingly by dividing by $\cos\varphi$ to give values corresponding to $\varphi=0°$.

and

$$n_A=-\sum_{i\uparrow} n_i^e(\uparrow)-\sum_{i\downarrow} n_i^e(\downarrow)+\sum_{j\uparrow} n_j^h(\uparrow)+\sum_{j\downarrow} n_j^h(\downarrow)z\,. \quad (9)$$

Adding Eqs. (8) and (9) we obtain

$$-n_A=sZ-[F(\uparrow)+F(\downarrow)+J(\uparrow)+J(\downarrow)], \quad (10)$$

where $F(\uparrow)$, $F(\downarrow)$ are the numbers of full spin zones and $J(\uparrow)$, $J(\downarrow)$ are the numbers of spin zones containing hole sheets of opposite signs of spin.

The important distinction between Eq. (4) for a nonmagnetic metal and Eq. (10) for a ferromagnetic metal is that (n_A+sZ) must be an *even* integer for the former, while this sum can also be an *odd* integer for the latter. This means that a ferromagnetic metal for which sZ is *odd* can be *compensated* ($n_A=0$), unlike a nonmagnetic metal. And a ferromagnetic metal for which sZ is *even* can be *uncompensated* with n_A equal to *any* positive or negative integer (in particular, $n_A=\pm1$), unlike a nonmagnetic metal.

IV. COMPENSATION IN FERROMAGNETIC METALS. EXPERIMENT AND INTERPRETATION

A. Nickel. Experiment

The anisotropy and field dependence of the magneto-resistance in applied fields up to 18 kOe for a high-purity nickel single crystal have been described previously.[1] The magnetoresistance saturates with increasing field for a general field direction. But it increases more rapidly than linearly when the field is near a symmetry axis or a symmetry plane due to the effects on the conductivity tensor of open cyclotron orbits. These measurements have been extended, and the field dependence of the Hall voltage has also been measured, up to an applied field of 80 kOe.[2] We shall be concerned here with the field dependence of the Hall voltage and the magnetoresistance for a general field direction, which are illustrated in Fig. 1. Above the ferromagnetic saturation field intensity $4\pi M$ of about 6 kOe the Hall voltage is a linear function of the applied field H, while the magnetoresistance continues to saturate up to 80 kOe. The complicated behavior of the magnetoresistance below 6 kOe is probably a consequence of the magnetocrystalline anisotropy, and is not relevant to the present paper. Also, one would expect the Hall voltage to be linear in the magnetic induction [$B=\mu_0(H+2\pi M)$ for H transverse to an infinite circular cylinder] to rather lower fields, but the estimation of B is difficult because of the magnetocrystalline anisotropy and the fact that the sample is of finite length. An accurate value of the Hall coefficient A_H above 6 kOe can be obtained just as well from a plot of voltage versus H.

The linear least-squares fit to the field dependence of the Hall voltage has a slope corresponding to $n_A=-1.06\pm0.03$ electrons per atom [$A_H=-6.4\times10^{-4}$ emu in Eq. (3)]. The probable error is estimated from the least-squares analysis and the internal consistency with measurements of A_H in other field directions. However, there may be a systematic error in all the measurements of A_H for the following reason: The sample was prepared "as-grown" from an electron beam melted rod, and probes for the Hall voltage measurements were soldered near the center of the sample so as to form a set of mutually perpendicular axes with the z axis of the rod. The separations d_x and d_y of the two pairs of probes were 2.200 and 2.217 mm, and in evaluating the current density from the measured current the cross-sectional area of the sample was estimated to be $\pi d_x d_y/4$. A systematic error would arise if the true cross-sectional area differed significantly from this value, as might easily happen due to an irregularity of the surface of the rod near the probes. It is considered that this error is unlikely to be as high as 10%, so the integer nearest the observed $n_A=-1.06$ is quite unambiguously minus one.

B. Nickel. Interpretation

The saturation of the magnetoresistance and the linearity of the Hall voltage with a value of A_H corresponding to one electron per atom are characteristic of an uncompensated metal. However, nickel with one atom per unit cell ($s=1$ for a fcc lattice) and an even atomic number ($Z=28$) will have an even value of N according to Eq. (7). It should, therefore, be compensated, by analogy with the nonmagnetic metals listed in Table I, or at least have an even-integral value of n_A.

The proposed explanation of this result at the time the measurements up to 18 kOe were published[1] was that the d band holes, equal in number to the s-band elec-

trons,[27] were of much lower mobility. According to this explanation the electrons are in the high-field region at 18 kOe,[28] while $\omega_c\bar{\tau}\ll 1$ for the holes at this field, and the latter therefore make a negligible contribution to the Hall term in the conductivity tensor. But in view of the continuing saturation of the magnetoresistance up to 80 kOe this explanation now seems most unlikely. A mobility ratio as low as 1/100 would still lead to a quadratic component of the magnetoresistance at 80 kOe roughly equal to the observed saturating component.[29] Though the high electronic specific heat of nickel suggests that the effective mass in the d band is about an order of magnitude greater than in the s band, there is no reason to suppose the impurity-scattering relaxation time also to be an order of magnitude shorter than for the s band electrons of the same sign of spin.

The resolution of the spin degeneracy of itinerant electrons in a ferromagnetic metal offers a more natural and convincing explanation of the observed behavior. This can be illustrated by means of the schematic band structure of nickel shown in Fig. 2. All but one branch of the d band are assumed to be below the Fermi level. The energy E for this branch of the d band, and for the s band, is shown as a function of the wave vector k along the line from the center Γ to the point X (arbitrarily chosen) on the cube face of the Brillouin zone.

Without the exchange interaction the number of electrons $n_s{}^e$ in the s band within an electron sheet centered on Γ must equal the number of holes in the d band within a hole sheet centered on X and its equivalent points. The exchange interaction is assumed to split the s band by a negligibly small amount, so the topologies of the spin-up sheets retain the electron character of the unperturbed sheet and the total number of electrons in the s band is essentially unchanged from $n_s{}^e$. But the exchange interaction splits the d band by a larger amount, and if we suppose that the spin-up (higher energy) sheet assumes an electron character while the spin-down sheet still intersects the Fermi level and retains its hole character, as shown in Fig. 2, we obtain the following equations:

$$n_s{}^e=1-n_d{}^e(\uparrow)+n_d{}^h(\downarrow), \tag{11}$$

and

$$-n_A-=n_s{}^e+n_d{}^e(\uparrow)-n_d{}^h(\downarrow). \tag{12}$$

Adding Eqs. (11) and (12), we obtain the required result, $n_A=-1$. This means physically that this model of nickel will exhibit a saturating magnetoresistance and a linear Hall voltage corresponding to one electron per atom for fields sufficiently high that all sheets of the Fermi surface are in the high-field region.

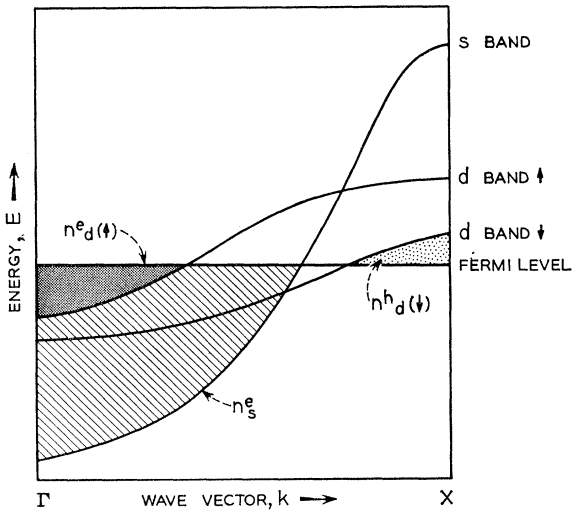

FIG. 2. Schematic band structure of nickel.

Of course, the schematic band structure of Fig. 2 is a gross simplification of the true band structure of nickel, in which more than one branch of the d band probably intersects the Fermi level even before the exchange splitting is introduced.[30] But a consideration of Eq. (10) shows that even so the observed behavior can be obtained if, when the exchange splitting is introduced in the d bands, the hole sheet in *one spin zone and one only* assumes an electron character. This follows from the assumptions that the electron characters of both the spin-up and spin-down sheets in the two spin zones of the s band are preserved and that there are no occupied states in bands derived from atomic states of higher quantum number. Thus, in Eq. (10), $F(\uparrow)+F(\downarrow)+J(\uparrow)+J(\downarrow)=27$, and since there is a total of 30 spin zones available, of which the two in the s band contain electron sheets, the sheet in the remaining spin zone in the d band must also have an electron character. Furthermore, topological considerations require that this should be a spin-up (higher energy) spin zone.

An interesting corollary of the conclusion that there must be an electron sheet in the d band is that this may be the multiply-connected sheet supporting open cyclotron orbits, rather than an electron sheet in the s band, as was suggested earlier.[1] There is no direct way to distinguish between these two possibilities from the galvanomagnetic measurements alone, though there is some evidence that the multiply connected sheet is of one sign of spin only.[2] The size of the "necks" near the point of contact L at the center of the hexagonal face of the Brillouin zone can be estimated from the angular widths of the magnetoresistance peaks caused by open cyclotron orbits running through the necks. An inde-

[27] The number of s-band electrons, $n_A=-0.82$, quoted in this paper was incorrect, since A_H was calculated from the Hall voltage per unit current density measured at an applied field $H=18$ kOe by dividing by H rather than B.

[28] The onset of saturation of the magnetoresistance in a noncompensated metal is a rough measure of the field at which $\omega_c\bar{\tau}=1$. For nickel this field is difficult to estimate because of the complicated behavior of the magnetoresistance at low fields, but the magnetoresistance curve shown in Fig. 1 suggests a value $\sim$10 kOe.

[29] This estimate is based on a simple two-band model, for which $\Delta\rho/\rho_0=(\omega_c\bar{\tau})_e(\omega_c\bar{\tau})_h$, with $(\omega_c\bar{\tau})_e\sim 8$ at 80 kOe.

[30] J. G. Hanus, MIT Solid State and Molecular Theory Group Quarterly Progr. Rept. No. 44, p. 29 (unpublished).

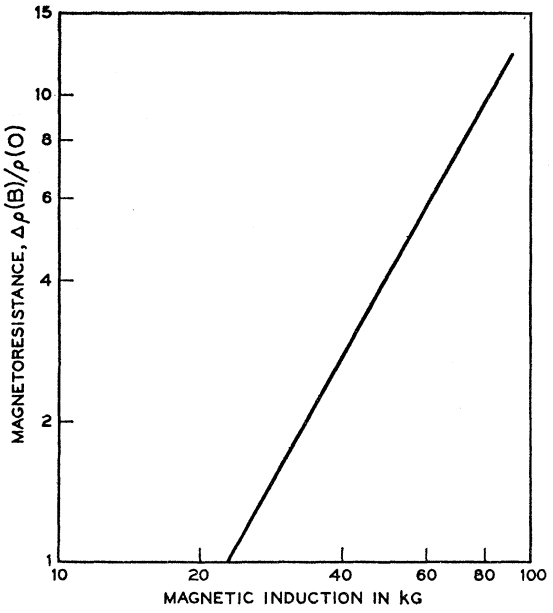

FIG. 3. Magnetoresistance of iron. The resistance ratio, $R_{295°K}/R_{4.2°K}$, of the sample in zero applied field was 191. The magnetic induction B is taken to be $\mu_0(H+2\pi M)$ for an applied field H transverse to a cylindrical sample, with $2\pi M=11$ kOe for iron.

pendent estimate of the size of the necks can be obtained from the observed increases in the Hall coefficient for the field along the symmetry axes over its value for a general field direction.[31] This estimate agrees somewhat better with that from the magnetoresistance peaks if the sheet is assumed to occupy a spin zone, accommodating one electron per atom, rather than a Brillouin zone. But these preliminary results are not conclusive, and further investigations on higher purity samples are needed. This example illustrates the importance of distinguishing between the spin zone and the Brillouin zone in deducing geometrical features of the Fermi surface from measurements of the Hall effect in ferromagnetic metals.

Ehrenreich and Phillip[32] have proposed a model of the band structure of nickel which explains its optical properties and is consistent with other experimental data, including the galvanomagnetic properties. They suggest that the exchange splitting in the s band is smaller than in the d bands, but is not negligible as we have assumed in Fig. 2. The spin-down (lower energy) sheet derived from the s band is multiply connected, while the spin-up sheet retains its electron character and is simply connected but is d-like rather than s-like over much of its surface. The third electron sheet is a spin-up sheet derived from a d band, as predicted from the galvanomagnetic properties.

[31] The fact that the Hall coefficient becomes *more* negative with the field along the symmetry axes shows that the multiply connected sheet is certainly an *electron* sheet, since this corresponds to hole orbits arising at the expense of electron orbits for these directions.

[32] H. Ehrenreich and H. R. Phillip, Bull. Am. Phys. Soc. 8, 257 (1963); and H. R. Phillip and H. Ehrenreich, Phys. Rev. **131**, 2016 (1963).

C. Iron. Experiment and Interpretation

The field dependence of the magnetoresistance of a single crystal of bcc iron of random orientation, with the transverse magnetic field in a general direction, is shown in Fig. 3. The transverse magnetoresistance in an applied field of 80 kOe was isotropic to within 10%, and its field dependence was everywhere similar.

The magnetoresistance shows no sign of saturation, and it seems likely that in a purer sample at higher fields it would approach even closer to a quadratic field dependence, characteristic of a compensated metal. One's confidence in this expcetation is enhanced by the empirical observation that, in sufficiently high magnetic fields, all the metals listed in Table I (and nickel) either approach a quadratic field dependence or saturate at a value less than 5, while the magnetoresistance of this sample of iron is already about 12 at an applied field of 80 kOe.

The conclusion that iron is compensated is not too informative about its band structure, but it is of course consistent with Eqs. (6) and (10), and indicates that the total number of full spin zones (outside the closed $3p$ shell) and spin zones containing hole sheets or either spin is equal to the maximum valency 8. Moreover, the result shows that, in this ferromagnetic metal at least, the mobility ratio between different bands cannot be significantly greater than in nonmagnetic metals.

V. CONCLUSION

We have shown quite generally that, in a metal in which the spin degeneracy of itinerant electrons is resolved by some interaction, the galvanomagnetic properties in the high-field region may be drastically affected. The difference from the nonmagnetic state is only manifested if an odd number of spin zones are full or contain hole sheets of the Fermi surface. This is only likely to happen in a ferromagnetic metal, where the energy splitting due to the ferromagnetic exchange interaction is comparable in magnitude to the widths of energy bands. But it may happen also in an antiferromagnetic metal, in which otherwise one expects compensation since the unit cell of the magnetic superlattice normally contains an even number of atoms.

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