Transport Theory for Ferromagnets*

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A quasiclassical transport theory for ferromagnets of large bandwidth is outlined, covering both the ordinary galvanomagnetic effects associated with the Lorentz force and the "spontaneous" effects related specifically to the magnetization. The latter effects are mediated by the spin-orbit interaction and are shown to consist of a scattering-independent Hall effect with no concurrent changes of the longitudinal resistivity, and a Hall effect accompanied by changes of this resistivity and which appears only in the presence of impurity scattering. The manner in which these various effects combine with the usual galvanomagnetic effects is also investigated.

I. INTRODUCTION

In a ferromagnetic metal, the galvanomagnetic effects appear to arise from two distinct physical phenomena. The first of these is a consequence of the ordinary Lorentz force due to the total magnetic induction field, while the second is related specifically to the magnetization and the corresponding effects are usually referred to as the "extraordinary," "anomalous," or "spontaneous" component. The exact nature of the relation with the magnetization and the way in which the ordinary and the spontaneous components combine to produce the observed galvanomagnetic effects will be examined in this paper.

According to the pioneering work of Karplus and Luttinger and of Smit,[1] the spontaneous effects are mediated by the spin-orbit interaction which introduces spin-dependent corrections in the expectation value of the position and velocity operators, via the off-diagonal part of these operators which depends on the spatial part of the wave functions. Some of the consequences of these corrections on charge transport have been investigated by Luttinger[2] and Adams and Blount[3] using full quantum-mechanical methods. These treatments differ by the basis functions chosen for the expansion of the density matrix, but they both indicate that if time-reversal symmetry is absent and conduction bands are coupled by spin-orbit interaction, two specific asymmetries show up in the transport properties and give rise in particular to a spontaneous Hall effect. Firstly, the changes in the velocities result in a transverse current independent of the scattering, and secondly, the scattering matrix becomes anisotropic as the scattering potential is modified by the spin-dependent changes of the position expectation value.

These qualitative results were important in settling a controversy between the former authors about the consistent accounting of all relevant terms in the density matrix, but the complexity of the formalism prevented an evaluation of the magnitudes expected for the magnetization-dependent currents. For this reason, the experimental data on the spontaneous effects have not yet received a satisfactory interpretation, although they appear to contain significant information about the Fermi surfaces and the mean free paths in the ferromagnetic metals. Among such data can be cited the deviations from Kohler's rule in Ni,[4] and other anomalies in Ni alloys,[5] the resistivity changes with intensity and direction of the magnetization, which are particularly large in Fe,[6] the giant Hall effects in Fe,[7] etc.

As a basis for the interpretation of these data, we outline in this paper a simple quasiclassical transport theory for ferromagnets of large bandwidth which aims at closed and directly usable formulas for the transport coefficients. In this theory, the Hamiltonian of the magnetized dissipative system is expanded in the crystal-momentum representation already used by Adams and Blount,[3] but with a particular choice of the phase of the wave functions which considerably simplifies the transport problem. In this way, the quasiclassical approach can be shown directly to describe the spontaneous effects with the same accuracy as the other ordinary effects, and the results so obtained are equivalent to the lowest-order results derived from full quantum calculations. The quasiclassical approach has, of course, the additional advantage of allowing relatively straightforward physical interpretations to be made, and since both spontaneous and ordinary effects are treated on the same footing, the manner in which they combine in the overall conductivity matrix for the ferromagnet can be easily established. In particular, it is shown that the rule usually assumed for the separation of the ordinary and spontaneous Hall effects is valid only in the small-field limit.

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II. HAMILTONIAN OF THE PROBLEM

We consider a ferromagnetic system described by the translationally invariant one-electron Hamiltonian:

$$H_p = H_0 + H_f + H_{so}, \tag{1}$$

where

$$H_0 = p^2/2m + \sum_n U(\mathbf{r}-\mathbf{n}) \tag{2}$$

is the usual band Hamiltonian containing a superposition of atomic potentials $U(\mathbf{r})$ at sites $\mathbf{n}$, and H_f is an additional term responsible for the splitting of the spin-degenerate bands of H_0 into up- and down-spin subbands. The spin-orbit term

$$H_{so} = \frac{-\hbar}{4m^2c^2} \sum_n \boldsymbol{\sigma}\cdot[\mathrm{grad}U(\mathbf{r}-\mathbf{n})\times\mathbf{p}] \tag{3}$$

is assumed to have small effects which can legitimately be investigated by a second-order perturbation theory applied to the unperturbed states of H_0+H_f. To this system is added some small external potential $V(\mathbf{r})$, representing impurities, phonons, etc., which can couple the states of H_p and induce irreversible processes which balance the driving force exerted by an applied electric field $\mathbf{E}$. Our problem is to evaluate the linear admittance of such a system, i.e., to consider a particle described by the Hamiltonian

$$H = H_p + V(\mathbf{r}) - e\mathbf{E}\cdot\mathbf{r}, \tag{4}$$

and to evaluate the total electric current

$$\mathbf{j} = (e/\hbar)\ \mathrm{Tr}\{\rho[H,\mathbf{r}]\} \tag{5}$$

to first order in the applied field, where ρ is the density matrix.

In order to make full use of the translational invariance of the system, we first expand both H_p and $\mathbf{r}$ in the so-called "crystal-momentum representation" which has already been discussed by Blount[8] and used extensively by Adams and Blount[3] in connection with this problem. In short, this representation is obtained from the energy representation of H_p by a cannonical transformation having the property of removing all the interband matrix elements of $\mathbf{r}$ but otherwise leaving its diagonal parts invariant to first order in the perturbation $V(\mathbf{r})-e\mathbf{E}\cdot\mathbf{r}$. This procedure provides a formal proof that a linear admittance as defined by Eq. (5) can be obtained correctly by considering H_p in Eq. (4) as a single band Hamiltonian. However, the price to pay for the isolation of the band is that the components of $\mathbf{r}$ do not necessarily commute in the new representation.

To calculate the commutator of the components of $\mathbf{r}$ for a particular isolated band, we use the crystal-momentum representation $\{\mathbf{k}\}$ generated by the set of Bloch functions pertaining to this band (band index omitted for brevity):

$$\langle\mathbf{r}|\mathbf{k}\rangle = u_k(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{6}$$

and obtain, for the matrix elements of $\mathbf{r}$ in this representation,

$$\langle\mathbf{k}'|\mathbf{r}|\mathbf{k}\rangle = \delta_{kk'}[\langle\mathbf{k}|\,\partial/i\partial\mathbf{k}|\mathbf{k}\rangle - \mathbf{J}(\mathbf{k})], \tag{7a}$$

with $\mathbf{J}(\mathbf{k})$ the vector defined by

$$\mathbf{J}(\mathbf{k}) = \int u_k^*(\mathbf{r})\frac{\partial u_k(\mathbf{r})}{i\partial\mathbf{k}}d\mathbf{r}. \tag{7b}$$

A difficulty has been generally recognized to arise at this point: While the left-hand side of Eq. (7a) is obviously phase-invariant, the two terms on the right-hand side depend on the phase of the wave functions; for instance, when u is changed to $u\times e^{i\phi(\mathbf{k})}$, $\mathbf{J}$ is changed to $\mathbf{J}+i\,\mathrm{grad}_k\phi(\mathbf{k})$. Instead of making a painstaking phase-invariant theory, we prefer here to make a definite choice of phases and show that this choice is always possible. As a matter of fact, the property of phase invariance of $\mathbf{r}$ has the same formal aspect as the property of gauge invariance of the velocity in quantum mechanics. Indeed, the velocity in the coordinate representation is a sum of two gauge-dependent terms:

$$\mathbf{v} = \frac{1}{m}\left(\frac{\hbar}{i}\frac{\partial}{\partial\mathbf{r}} - \frac{e}{c}\mathbf{A}(\mathbf{r})\right), \tag{8a}$$

where $\mathbf{A}(\mathbf{r})$ is the vector potential, but the commutator of the components of $\mathbf{v}$,

$$[\mathbf{v},\mathbf{v}] = (e/c)\ \mathrm{curl}\mathbf{A}(\mathbf{r}), \tag{8b}$$

does not vanish in general. The formal similarity with our problem, which also exhibits these two features, arises because a change of the electromagnetic gauge is in fact equivalent to a change of the phase factor in the wave functions. Since we know from the electromagnetic problem that the above commutator is gauge-invariant and that a useful choice for the gauge is the radiation gauge $\mathrm{div}\mathbf{A}(\mathbf{r})=0$, we deduce that $\mathbf{r}$ can have the same formal properties in the representation $\{\mathbf{k}\}$, namely, the commutator

$$[\mathbf{r},\mathbf{r}] = \mathrm{curl}_k\mathbf{J}(\mathbf{k}) \tag{9a}$$

is a phase-invariant quantity, and

$$\mathrm{div}_k\mathbf{J}(\mathbf{k}) = 0 \tag{9b}$$

is a convenient choice for the phases of the Bloch functions in Eq. (6).

To show that the choice in Eq. (9b) is always possible, we use the following geometrical interpretation, which is based on the observation that no physically significant meaning is attached to the origin of $\mathbf{r}$ in a translationally invariant medium such as a crystal. This origin may therefore be selected to our best advantage so long as any translation (possibly different for each $\mathbf{k}$) considered for this origin affects the wave functions only through their (possibly $\mathbf{k}$-dependent) phase factor. This arbi-

[8] E. I. Blount, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1962), Vol. 13, p. 305.

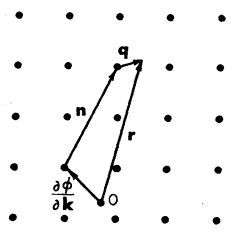

FIG. 1. Separation of the coordinate vector **r**, or origin *O*, according to the relation in Eq. (9) of the text.

trariness of the origin can be represented by the formal separation illustrated for a square lattice by Fig. 1:

$$\mathbf{r}=\mathbf{n}+\mathbf{q}+\mathrm{grad}_k\phi(\mathbf{k}), \tag{10}$$

where **n** is a lattice vector, **q** is a translationally invariant vector field with its origin at the center of a cell, and $\phi(\mathbf{k})$ is an arbitrary scalar function of **k** which can always be accepted as a phase factor in Eq. (7).

As an operator equation, the relation (10) immediately lends itself to the cellular operator method originally developed by Wannier.[9] In this method, **n** and **k** are considered as conjugate variables which are diagonal in the {**n**} and {**k**} representations generated by the Wannier and Block functions, respectively, and, as emphasized by Blount, these two representations are in the same relationship as the coordinate and momentum representations for the ordinary Schrödinger equation. Therefore transformations from one to the other do not involve any approximation, if any function of these variables is defined only at their discrete allowed values in the crystal. Since this limitation is of little practical importance in a large crystal, we will henceforth consider **n** and **k** to be continuous conjugate variables with which are associated the following pairs of noncommuting operators:

$$\mathbf{n},\mathbf{k}=-i\partial/\partial\mathbf{n}; \quad \mathbf{k},\mathbf{n}=-i\partial/\partial\mathbf{k}, \tag{11}$$

in the {**n**} and {**k**} representations, respectively.

Using Eq. (11), we obtain for Eq. (10) an operator relation whose general validity and significance has been discussed by Zak.[10] For our purpose, it will be sufficient to consider Eq. (10) in the {**k**} representation where the translationally invariant operator **q** has an expectation value $\langle\mathbf{k}|\mathbf{q}|\mathbf{k}\rangle$, also abbreviated as $\langle\mathbf{q}(\mathbf{k})\rangle$, depending only on **k**. We thus easily verify the relation (9) as well as its phase invariance and from Eq. (7), we find that $\mathbf{J}(\mathbf{k})$ has zero divergence as proposed in Eq. (9b), if the Poisson equation

$$\nabla_k^2\phi(\mathbf{k})=-\mathrm{div}_k\langle\mathbf{q}(\mathbf{k})\rangle \tag{12}$$

is satisfied. Since this equation can always be solved, we conclude that the desired choice of phases is always possible; in fact, our argument is completely analogous to the usual demonstration of the existence of the radiation gauge.[11]

[9] G. H. Wannier, in *Elements of Solid State Theory* (Cambridge University Press, Cambridge, England, 1959), Chap. 6.
[10] J. Zak, Phys. Rev. Letters **19**, 1385 (1967).
[11] See, e.g., D. Bohm, in *Quantum Theory* (Constable Co., Inc., London, 1954), pp. 7 and 358.

The choice of phases $\mathrm{div}_k\mathbf{J}(\mathbf{k})=0$ has already been suggested by Blount[8] on the ground that it minimizes the spread of the Wannier functions obtained from Eq. (6). Decomposing the **q** field into an irrotational component $\mathbf{q}_l(\mathrm{curl}_k\mathbf{q}_l=0)$ and a solenoidal component $\mathbf{q}_r(\mathrm{div}_k\mathbf{q}_r=0)$, we find in addition that this choice of phases removes the irrotational part $\mathbf{q}_l$, leaving only

$$\mathbf{r}=\mathbf{n}+\mathbf{q}_r, \tag{13}$$

with the expectation value of $\mathbf{q}_r$ given by

$$\langle\mathbf{q}_r(\mathbf{k})\rangle=\frac{1}{4\pi}\int_{\mathrm{BZ}}\mathrm{curl}_{k'}\langle\mathbf{q}(\mathbf{k}')\rangle\times\frac{\mathbf{k}-\mathbf{k}'}{|\mathbf{k}-\mathbf{k}'|^3}d\mathbf{k}', \tag{14}$$

where the integration extends over the primitive Brillouin zone (BZ). The vector $\langle\mathbf{q}_r\rangle$, which we will call the "solenoidal polarizability," is a periodic function of **k** only and is defined within an unimportant lattice vector. In presence of a center of spherical symmetry, integration of (14) over the angle θ between **k** and **k**′ gives

$$\begin{aligned}\langle\mathbf{q}_r(\mathbf{k})\rangle&=\frac{1}{4\pi}\int\left(\mathrm{curl}_{k'}\langle\mathbf{q}(\mathbf{k}')\rangle\times\frac{\mathbf{k}}{k}\right)\frac{k-k'\cos\theta}{|\mathbf{k}-\mathbf{k}'|^3}d\mathbf{k}'\\&=\int_0^k\left[\mathrm{curl}_{k'}\langle\mathbf{q}(\mathbf{k}')\rangle\times\mathbf{k}\right]\frac{k'^2dk'}{k^3},\end{aligned} \tag{15}$$

since the integrand vanishes identically for $k'>k$.

Finally, we rewrite the original Hamiltonian of the crystal in the electric field by substituting Eq. (13) into Eq. (4) and find

$$H=H_p(\mathbf{k})-e\mathbf{E}\cdot\mathbf{q}_r(\mathbf{k})+V[\mathbf{n}+\mathbf{q}_r(\mathbf{k})]-e\mathbf{E}\cdot\mathbf{n}, \tag{16}$$

for which we can seek solutions either in the {**n**} representation or in the {**k**} representation according to the circumstances, and such solutions represent the solutions of Eq. (4) in the form of admixtures of Wannier or Bloch functions, respectively. Since in this paper we are particularly interested in the effects of continuous perturbations which vary slowly in space, it is appropriate to consider Eq. (16) in the {**n**} representation, where it represents a particle of coordinate **n** moving with some "kinetic energy" H_p and subjected to various coordinate and momentum-dependent potentials, in addition to the electric driving term $-e\mathbf{E}\cdot\mathbf{n}$. We thus have converted the transport problem in a crystal into the comparatively simple problem of an equivalent particle moving in the same electric field in ordinary space. No major quantum effects should arise in such a situation, and quasiclassical methods are presumably adequate to evaluate the transport effects. We shall see that such an approach can be used with advantage in a ferromagnet, provided that simple expressions are obtained for the "kinetic" term and the momentum-dependent potentials.

Actually, the above discussion is, in principle, valid for any kind of crystal and is often used to show the

effective-mass theorem in a shorter version overlooking the vector field $\mathbf{q}$ altogether. This is a legitimate step if H_p has time-reversal invariance because, even though the spin-orbit interaction may make $\mathbf{q}$ dependent on the spin as shown below, the two degenerate spin states give opposite contributions which cancel each other. Therefore, the $\mathbf{q}$ field can give rise to significant physical effects only if the populations of up- and down-spin states differ, e.g., in a ferromagnet.

The field $\mathbf{q}$ is easily evaluated by considering H_{so} in Eq. (1) as a perturbation on H_0+H_f and coupling states of identical $\mathbf{k}$. In order to include the important case of accidental degeneracies in the simplest manner, we consider here only the coupling between a pair of bands α and α', as illustrated in Fig. 2. With ω the energy separation between the unperturbed states, and

$$\tfrac{1}{2}\Delta(\mathbf{k})=\langle\mathbf{k}_{\alpha'}|H_{so}|\mathbf{k}_{\alpha}\rangle, \tag{17}$$

the interband matrix element of H_{so}, a second-order perturbation calculation yields the energy shifts

$$\delta\epsilon=\pm\tfrac{1}{2}|\Delta|^2/(\omega^2+|\Delta|^2)^{1/2}. \tag{18}$$

The corresponding modifications to the velocities are given by

$$\delta\mathbf{v}=\pm\frac{1}{4}\frac{2\omega^2+|\Delta|^2}{(\omega^2+|\Delta|^2)^{3/2}}\left(\frac{\Delta^*d\Delta}{d\hbar\mathbf{k}}+\frac{\Delta d\Delta^*}{d\hbar\mathbf{k}}\right)\mp\frac{\omega|\Delta|^2}{(\omega^2+|\Delta|^2)^{3/2}}\frac{d\omega}{d\hbar\mathbf{k}}, \tag{19}$$

and the expectation value of $\mathbf{q}$ is

$$\langle\mathbf{q}\rangle=\pm[\tfrac{1}{2}\Delta/(\omega^2+|\Delta|^2)^{1/2}]\langle\mathbf{k}_{\alpha}|\mathbf{q}|\mathbf{k}_{\alpha}'\rangle. \tag{20}$$

In the last three expressions, the upper and lower signs refer to the states shifted upwards or downwards, respectively, by the spin-orbit interaction.

All the above changes induced by the spin-orbit interaction may produce observable effects in the transport properties. If the Fermi level happens to be close to a degeneracy, which appears to be a common feature of the ferromagnetic metals, the first-order energy shifts in Eq. (18) induce spin-dependent changes in occupancy, i.e., in the shape of the Fermi surface. The associated modifications of the velocity field in Eq. (19) add effects which are even in the spins, as noted by Karplus and Luttinger, and may give rise to an anisotropy of the resistance of ferromagnetic origin. Finally, the effects connected with the polarizability $\langle\mathbf{q}\rangle$, i.e., with the driving and scattering potentials, are restricted to the coupling of bands of the *same* spin only, since the operator $\mathbf{q}$ is spin-independent; they can be even in the magnetization and thus add to the ferromagnetic anisotropy of the resistance, or they can be odd in the magnetization and are then measured as the spontaneous Hall effect.

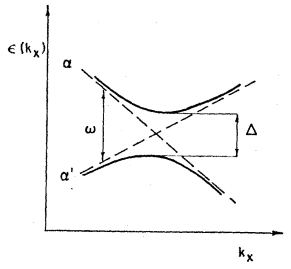

FIG. 2. Energy diagram for a pair of bands α and α' coupled by the spin-orbit interaction.

III. EFFECTIVE HAMILTONIAN

Since we want to deal with limited regions of $\mathbf{k}$ space around points $\mathbf{k}_i$ of special interest, we can assume that an orthonormal cellular basis $|\mathbf{n}_\alpha\rangle$ of the Wannier type exists such that the Bloch sums

$$|\mathbf{k}_\alpha\rangle=\sum_n e^{-i\mathbf{k}\cdot\mathbf{n}}|\mathbf{n}_\alpha\rangle/\sqrt{N} \tag{21}$$

diagonalize approximately the Hamiltonian H_0+H_f for $\mathbf{k}-\mathbf{k}_i$ small. Since H_0+H_f has the full cubic symmetry, including inversion symmetry, we have

$$\epsilon(\mathbf{k})=\langle\mathbf{0}_\alpha|H_0+H_f|\mathbf{0}_\alpha\rangle+\sum_{\rho\neq0}\langle\mathbf{0}_\alpha|H_0+H_f|\boldsymbol{\rho}_\alpha\rangle\cos\mathbf{k}\cdot\boldsymbol{\rho}, \tag{22}$$

$$\mathbf{v}(\mathbf{k})=-\hbar^{-1}\sum_{\rho\neq0}\langle\mathbf{0}_\alpha|H_0+H_f|\boldsymbol{\rho}_\alpha\rangle\sin\mathbf{k}\cdot\boldsymbol{\rho}.$$

In the nearest-neighbor approximation, we can write

$$\mathbf{v}(\mathbf{k})=-\frac{\Delta\epsilon}{2\hbar}\sum_\rho\boldsymbol{\rho}\sin\mathbf{k}\cdot\boldsymbol{\rho}, \tag{23}$$

where $\Delta\epsilon=2\langle\mathbf{0}|H_0+H_f|\boldsymbol{\rho}\rangle$ is the appropriate bandwidth. A similar expansion can be made for H_{so} since it also has inversion invariance, but only the intracell element needs to be retained since the intercell elements can be shown to be very small because of the orthogonality of the Wannier functions:

$$\tfrac{1}{2}\Delta(\mathbf{k})=\langle\mathbf{0}_{\alpha'}|(-\hbar\boldsymbol{\sigma}/4m^2c^2)(\nabla U(\mathbf{r})\times\mathbf{p})|\mathbf{0}_\alpha\rangle. \tag{24}$$

Since only the spherically symmetric part of $U(\mathbf{r})$ has a large gradient, we approximate

$$\Delta(\mathbf{k})=2\langle\mathbf{0}_{\alpha'}|\,\xi(r)\boldsymbol{\sigma}\cdot\mathbf{L}|\,\mathbf{0}_\alpha\rangle, \tag{25}$$

with $\mathbf{L}=\mathbf{r}\times\mathbf{p}$ and

$$\xi(r)=-\frac{\hbar}{4m^2c^2}\frac{\partial U}{r\partial r}. \tag{26}$$

As in interpolation schemes for transition metals,[12] we consider that an appropriate tight-binding basis $|\boldsymbol{\rho}^T\rangle$ can be formed with spherical atomiclike functions. To first order in the overlap between these functions, the intracell element in the Wannier basis and the tight-

[12] L. Hodges *et al.*, Phys. Rev. **152**, 505 (1966).

binding basis are identical and their radial and angular parts can be separated:

$$\Delta(\mathbf{k})=2\bar{\xi}\langle 0_{\alpha'}{}^{T}|\boldsymbol{\sigma}\cdot\mathbf{L}|0_{\alpha}{}^{T}\rangle, \tag{27}$$

with the relevant spin-orbit parameter defined by

$$\bar{\xi}=\langle 0_{\alpha'}{}^{T}|\xi(r)|0_{\alpha}{}^{T}\rangle. \tag{28}$$

By using the separation

$$\boldsymbol{\sigma}\cdot\mathbf{L}=\sigma_u L_u+\tfrac{1}{2}(\sigma_+ L_-+\sigma_- L_+), \tag{29}$$

the expectation value of the spin-orbit operator is readily evaluated for a spin quantization direction $\mathbf{u}$ parallel to the local magnetization. In this case, σ_u has the expectation values $s=+1$ and $s=-1$ for up- and down-spin electrons, respectively. Let $\mathbf{u}_1$ and $\mathbf{u}_2$ be two unit vectors such that $\mathbf{u}_1$, $\mathbf{u}_2$, and $\mathbf{u}$ form an orthogonal triad related to the unit vectors $\hat{x}$, $\hat{y}$, and $\hat{z}$ along the principal axes of the cubic structure by the rotation matrix:

$$\begin{bmatrix}\mathbf{u}_1\\ \mathbf{u}_2\\ \mathbf{u}\end{bmatrix}=\begin{bmatrix}\cos\theta\cos\phi & \cos\theta\sin\phi & -\sin\theta\\ -\sin\phi & \cos\theta & 0\\ \sin\theta\cos\phi & \sin\theta\sin\phi & \cos\theta\end{bmatrix}\begin{bmatrix}\hat{x}\\ \hat{y}\\ \hat{z}\end{bmatrix},$$

with θ and ϕ as the polar angles of $\mathbf{u}$ with respect to the cubic axes. One then finds from the rules for rotation of angular momenta that

$$\begin{aligned}\sigma_u L_u&=s\mathbf{u}\cdot\mathbf{L},\\ \sigma_+ L_-&=(\mathbf{u}_1-i\mathbf{u}_2)\cdot\mathbf{L},\\ \sigma_- L_+&=(\mathbf{u}_1+i\mathbf{u}_2)\cdot\mathbf{L}^*,\end{aligned} \tag{30}$$

where the vectorial operator $\mathbf{L}$ is defined by

$$\mathbf{L}=\hat{x}L_x+\hat{y}L_y+\hat{z}L_z.$$

Insertion of (29) and (30) into (27) gives matrix elements of the spin-orbit interaction which are independent of $\mathbf{k}$ and depend only on the angle between the magnetization direction and the expectation value $\langle\mathbf{L}\rangle=\langle 0_{\alpha'}{}^{T}|\mathbf{L}|0_{\alpha}{}^{T}\rangle$, which is a fixed vector characteristic of the pair of bands considered. In particular, the diagonal-in-spin element is

$$\Delta(\mathbf{k})=2\bar{\xi}(s\mathbf{u}\cdot\langle\mathbf{L}\rangle). \tag{31}$$

As $\mathbf{q}$ is odd under inversion, its expansion of the Wannier basis is

$$\langle\mathbf{k}_{\alpha'}|\mathbf{q}|\mathbf{k}_{\alpha}\rangle=i\sum_{\rho}\langle 0_{\alpha'}|\mathbf{q}|\boldsymbol{\varrho}_{\alpha}\rangle\sin\mathbf{k}\cdot\boldsymbol{\varrho}. \tag{32}$$

Using the translation operator $T_\rho=\exp(i\boldsymbol{\varrho}\cdot\mathbf{p}/\hbar)$, and summing over the two periods of $\mathbf{q}$ covering the states $|0\rangle$ and $|\boldsymbol{\varrho}\rangle$, we find

$$\begin{aligned}\langle 0|\mathbf{q}|\boldsymbol{\varrho}\rangle&=\langle 0|\mathbf{q}T_\rho|0\rangle+\langle 0|\tilde{T}_{-\rho}\mathbf{q}|0\rangle\\ &=2\langle 0|\mathbf{q}T_\rho|0\rangle+\langle 0|[T_\rho,\mathbf{q}]|0\rangle.\end{aligned} \tag{33}$$

For small overlap, $\mathbf{q}$ is practically identical to $\mathbf{r}$ wherever the Wannier functions have large gradients. Then the last term of Eq. (33) vanishes by orthogonality of the cellular functions, since

$$[T,\mathbf{r}]=-i\hbar\partial T/\partial\mathbf{p}=\boldsymbol{\varrho}T. \tag{34}$$

Moreover, only the odd terms in the expansion of the exponential contribute to the solenoidal component of the $\mathbf{q}$ field which corresponds to the magnetic dipole part of the operator ($i, j=x, y, z$)

$$q_i\nabla_j=\tfrac{1}{2}[(q_i\nabla_j+q_j\nabla_i)+(q_i\nabla_j-q_j\nabla_i)]. \tag{35}$$

Within the nearest-neighbor approximation (22), the solenoidal component can finally be written

$$\langle\mathbf{k}_{\alpha'}|\mathbf{q}|\mathbf{k}\rangle=(2\chi/\Delta\epsilon)(\mathbf{v}\times\langle\mathbf{L}\rangle), \tag{36}$$

with χ a constant of the order of the overlap.

Derivation of (19) with Δ constant yields

$$\begin{aligned}\operatorname{curl}\langle\mathbf{q}\rangle=&\pm[\tfrac{1}{2}\Delta/(\omega^2+|\Delta|^2)^{1/2}]\operatorname{curl}\langle\mathbf{k}_\alpha|\mathbf{q}|\mathbf{k}_{\alpha'}\rangle\\ &\mp[\Delta d\omega^2/d\mathbf{k}/(\omega^2+|\Delta|^2)^{3/2}]\times\langle\mathbf{k}_\alpha|\mathbf{q}|\mathbf{k}_{\alpha'}\rangle.\end{aligned} \tag{37}$$

Near the center of the Brilloin zone, around which an approximate spherical symmetry exists in general, only the first term of Eq. (37) is of zero order in $\mathbf{k}$ and we can use Eq. (15) to evaluate the polarizability. Inserting Eqs. (23), (25), and (35), we obtain for a pair of nondegenerate bands

$$\langle\mathbf{q}_r(\mathbf{k})\rangle=-(\bar{\xi}\rho^2/\omega)\chi[s(\mathbf{u}\cdot\langle\mathbf{L}\rangle)(\langle\mathbf{L}\rangle^*\times\mathbf{k})]. \tag{38}$$

The accidental degeneracies, near which all quantities can be regarded as slowly varying by comparison with ω, are also points of nearly spherical symmetry. As such degeneracies occur simultaneously at all points corresponding to each other by the Brilloin-zone symmetry, the last term of Eq. (37) need not be considered, since its contributions are linear in $\langle\mathbf{k}_\alpha|\mathbf{q}|\mathbf{k}_{\alpha'}\rangle$ and therefore cancel each other by inversion symmetry when they are eventually summed over all corresponding points of degeneracy. Assuming that ω varies linearly with the distance to the point of degeneracy $\mathbf{k}_i$, we find for the first term when $\mathbf{k}-\mathbf{k}_i$ is small

$$\begin{aligned}\langle\mathbf{q}_r(\mathbf{k})\rangle=&\mp(\chi\bar{\xi}/\Delta\epsilon)\operatorname{div}\mathbf{v}_i\{s(\mathbf{u}\cdot\langle\mathbf{L}\rangle)[\langle\mathbf{L}\rangle^*\times(\mathbf{k}-\mathbf{k}_i)]\}\\ &\times\frac{1}{\omega^3}\int_0^{\omega}\frac{\omega^2 d\omega}{(\omega^2+|\Delta|^2)^{1/2}}.\end{aligned} \tag{39}$$

Thus $\langle\mathbf{q}_r\rangle$ is of zero order in the spin-orbit parameter near the degeneracy, i.e., for $\omega<\Delta$,

$$\langle\mathbf{q}_r(\mathbf{k})\rangle=\mp\frac{\chi\operatorname{div}\mathbf{v}_i}{3\Delta\epsilon}\frac{(s\mathbf{u}\cdot\langle\mathbf{L}\rangle)}{|s\mathbf{u}\cdot\langle\mathbf{L}\rangle|}[\langle\mathbf{L}\rangle^*\times(\mathbf{k}-\mathbf{k}_i)]. \tag{40}$$

Away from the degeneracy and insofar as the linear relationship between ω and $|\mathbf{k}-\mathbf{k}_i|$ is valid, the polarizability decreases to a constant value

$$\langle\mathbf{q}_r(\mathbf{k})\rangle=\mp\frac{\operatorname{div}\mathbf{v}i}{2\Delta\epsilon}\chi\bar{\xi}(s\mathbf{u}\cdot\langle\mathbf{L}\rangle)\frac{\langle\mathbf{L}\rangle^*\times(\mathbf{k}-\mathbf{k}_i)}{\omega}, \tag{41}$$

which may be large if the crossing bands are flat, i.e., if the effective masses are large.

It is noteworthy that for any given pair of interacting bands, $\langle \mathbf{q}_r \rangle$ is not normal to the direction of magnetization $\mathbf{u}$, but to a direction specified by $\langle \mathbf{L} \rangle$ and which is rigid with respect to the crystal orientation. Therefore, transport effects connected with $\langle \mathbf{q} \rangle$ are intrinsically anisotropic, and large anisotropy can be expected when a particular pair of bands dominates over all others as in the case of degeneracies. However, in the general case that we will henceforth consider, several pairs of nondegenerate bands are involved, each with a vector $\langle \mathbf{L} \rangle$ differently oriented, so that only the components along the direction $\mathbf{u}$ add constructively in Eq. (38). Then an isotropic approximation is obtained in which the polarizability is described by

$$\langle \mathbf{q}_r(\mathbf{k}) \rangle = (\hbar \tau_q / m^*)(s\mathbf{u} \times \mathbf{k}), \tag{42}$$

$$\tau_q = (m^*/3\hbar) \sum (\chi \xi \rho^2/\omega) |\langle \mathbf{L} \rangle|^2, \tag{43}$$

where the summation sign indicates a summation over all bands interacting with the band considered.

In Eq. (42), a factor m^* has been introduced for convenience in reformulating the original Hamiltonian in the effective-mass approximation. Such an approximation, of course, is legitimate for evaluating the properties of the total Hamiltonian in Eq. (4) only if $V(\mathbf{r})$ is a slowly varying function, and we may accordingly use its Taylor expansion limited to the first-order term:

$$V(\mathbf{r}) = V(\mathbf{n}) + \mathbf{q}_r \cdot \mathrm{grad}_n V(\mathbf{n}). \tag{44}$$

Introducing Eqs. (42) and (44) into Eq. (16), we obtain the final Hamiltonian

$$H = H_{\mathrm{eff}} - e\mathbf{E} \cdot \mathbf{n}, \tag{45}$$

with an effective Hamiltonian for the dissipative system given by

$$H_{\mathrm{eff}} = \frac{\hbar^2 k^2}{2m^*} + \frac{e\hbar\tau_q}{m^*}(s\mathbf{u} \times \mathbf{E}) \cdot \mathbf{k} - \frac{\hbar \tau_q}{m^*} s\mathbf{u} \cdot [\mathrm{grad}_n V(\mathbf{n}) \times \mathbf{k}] + V(\mathbf{n}). \tag{46}$$

This Hamiltonian is readily recognized as representing a spinless particle of coordinate $\mathbf{n}$ and mass m^* which moves in free space under the combined influence of driving and scattering forces depending only on $\mathbf{n}$. There are, however, two additional momentum-dependent forces whose asymmetry gives rise to the spontaneous Hall effect as mentioned in the Introduction.

The formal structure of the momentum-dependent terms in Eq. (46) is seen to be identical with the spin-dependent terms derived from the Dirac equation, except that the spin operators are here replaced by ordinary numbers. Blount[8] and Adams and Blount[3] were the first to point out this far-reaching analogy which finds its origin in the fact that, like the crystal Hamiltonian, the Dirac Hamiltonian has a multiband spectrum of eigenvalues. In both situations, the set of eigenstates of a given band represents a particle of finite size, whose spatial extent is related to the energy gaps between the bands.[13] As a result, the interaction of that particle with external fields is nonlocal, and therefore depends also on the derivatives of the potential of the point charge. In their paper, Adams and Blount[3] emphasized the ensuing analogy between the "anomalous current" associated with the second term in Eq. (46) and the "spin current" which flows when a Dirac particle is accelerated. It is seen here that the analogy is readily extended to the third term in Eq. (46), which is the explicit analog of the Dirac's spin-orbit term and will be shown later to be responsible for the anisotropy of the scattering first proposed by Smit.

Both the second and third terms in Eq. (46) give contributions to the current, which are linear in the electric field and are analyzed separately in the next section.

IV. QUASICLASSICAL TRANSPORT THEORY

In a quasiclassical theory, the transport effects in a translationally invariant system with Hamiltonian H_p, eigenstates $|\mathbf{k}\rangle$ and eigenenergies ϵ_k, are described by means of a distribution function

$$f_k = f^0 + f_1(\mathbf{k}), \quad f_1(\mathbf{k}) = [\partial f_1(\mathbf{k})/\partial \mathbf{k}] \cdot \mathbf{k} \tag{47}$$

obeying the linearized Boltzmann equation

$$\frac{-e\mathbf{E}}{\hbar} \cdot \frac{\partial f^0}{\partial \mathbf{k}} = -\sum_{k'} (f_{k'} - f_k) Q_{kk'}, \tag{48}$$

with $Q_{kk'}$ the total transition probability from $|\mathbf{k}\rangle$ to $|\mathbf{k}'\rangle$. If this probability is assumed to depend only on the angle θ between $\mathbf{k}$ and $\mathbf{k}'$, the Boltzmann equation takes the form

$$\frac{-e\mathbf{E}}{\hbar} \cdot \frac{\partial f^0}{\partial \mathbf{k}} = \frac{\partial f_1}{\partial \mathbf{k}} \cdot \frac{\mathbf{k}}{\tau}, \tag{49}$$

with

$$\tau^{-1} = \int Q_{kk'}(1 - \cos\theta) \frac{d\mathbf{k}'}{8\pi^3}. \tag{50}$$

If a small magnetic induction field of direction $\mathbf{u}$ and intensity B is applied, an additional term accounts for the Lorentz force:

$$\frac{-e\mathbf{E}}{\hbar} \cdot \frac{\partial f^0}{\partial \mathbf{k}} = \frac{\partial f_1}{\partial \mathbf{k}} \cdot \left(\frac{\mathbf{k}}{\tau} + \frac{\mathbf{u} \times \mathbf{k}}{\tau_B} \right), \quad \tau_B^{-1} = \frac{eB}{m^*}. \tag{51}$$

This equation is easily solved for $\partial f_1/\partial \mathbf{k}$ and the current is found to have the general form

$$\mathbf{j} = -\tfrac{1}{3} e^2 \sum_k \tau v^2 \times \frac{df^0}{d\epsilon} \frac{\mathbf{E} + (\tau/\tau_B)^2 \mathbf{u}(\mathbf{u} \cdot \mathbf{E}) - (\tau/\tau_B)(\mathbf{u} \times \mathbf{E})}{1 + (\tau/\tau_B)^2}, \tag{52}$$

[13] A. Messiah, in *Mécanique Quantique II* (Dunod Cie., Paris, 1960), Chap. XX.

describing the well-known Hall and magnetoresistive effects due to the curving of the particle trajectories by the magnetic field.

The two terms in s in Eq. (46) are easily included in this framework as perturbations to the crystal Hamiltonian H_p. However, as they are not invariant under time reversal, they introduce corrections $\delta Q_{kk'}$ into the transition probabilities $Q_{kk'}$ violating the principle of microscopic reversibility on which the Boltzmann equation is based. In terms of the variational transport theory,[14] the corresponding additional scattering operators are not self-adjoint, a property shared by other forces linear in the velocity, such as the magnetic Lorentz force, and which do not contribute to the production of entropy. For such cases, it has been shown that the Boltzmann equation is nevertheless an acceptable solution for the variational principle of transport theory since it makes the entropy production at least stationary (instead of extremal with self-adjoint operators). We will therefore include the corrections $\delta Q_{kk'}$ in the Boltzmann equation without further justification.

A. Transverse Drift

For reasons apparent below, we will call "transverse drift" the transport effects associated with the second term in Eq. (46). Since this term is diagonal in the energy representation of H_p, it causes $\mathbf{k}$-dependent energy shifts and corresponding changes of the velocities given by

$$\delta\epsilon=(he\tau_q/m^*)(s\mathbf{u}\times\mathbf{E})\cdot\mathbf{k}, \qquad (53)$$
$$\delta\mathbf{v}=(e\tau_q/m^*)(s\mathbf{u}\times\mathbf{E}).$$

A possible interpretation of these shifts is that the spin-orbit interaction, as it transmits the time-reversal asymmetry of the spin part of the electron system to its spatial part, distorts the charge density and induces a dipole moment which then interacts with the electric field. The corresponding net current change is represented by

$$\delta\mathbf{j}=e\sum_k\delta(\mathbf{v}f)=e\sum_k\left(\mathbf{v}\frac{df^0}{d\epsilon}\delta\epsilon+f^0\frac{d\delta\epsilon}{d\hbar\mathbf{k}}\right)$$
$$=\frac{e}{\hbar}\frac{d}{d\mathbf{k}}\sum_k f^0\delta\epsilon=0, \qquad (54)$$

which vanishes because $\delta\epsilon$ is odd in $\mathbf{k}$. Thus, the current due to the variation of the velocities is exactly compensated by the depopulation caused by the simultaneous variation of the energy. However, as noted by Doniach,[15] the energy shifts also modify the scattering probabilities in Eq. (48). Within the Born approximation assumed in Eq. (49), the transition rate is thus changed by

$$\delta Q_{kk'}=Q_{kk'}(\delta\epsilon_k-\delta\epsilon_{k'})df^0/d\epsilon, \qquad (55)$$

which changes sign under interchange of $\mathbf{k}$ and $\mathbf{k}'$. As noted above, $Q_{kk'}$ does not contribute to the entropy production, but can still be introduced into the Boltzmann equation:

$$\frac{-e\mathbf{E}}{\hbar}\cdot\frac{\partial f^0}{\partial\mathbf{k}}=\frac{\partial f_1}{\partial\mathbf{k}}\cdot\frac{\mathbf{k}}{\tau}-\frac{he\tau_q}{m^*}\frac{df^0}{d\epsilon}(\mathbf{E}\times s\mathbf{u})\cdot\frac{\mathbf{k}}{\tau}. \qquad (56)$$

The additional term is structurally different from the Lorentz term in Eq. (51) and generates a transverse current given by

$$\delta\mathbf{j}=-(\mathbf{E}\times s\mathbf{u})\tfrac{1}{3}e^2\sum_k\frac{df^0}{d\epsilon}\tau_q v^2. \qquad (57)$$

This spontaneous current is independent of the scattering and results from a net average drift of the particles leaving the longitudinal current unchanged. Thus the apparent mean free path is paradoxically longer in the spin field, instead of being shorter as in a magnetic field which curves the trajectories.

Basically, the contribution in Eq. (57) represents a change of the scattering efficiency by which a particle having gained energy is allowed a shorter mean free time as more empty states are available for scattering at that higher energy. It therefore corresponds exactly to the scattering effects, which Smit claimed would cancel the anomalous contribution proposed by Karplus and Luttinger.[1] Since the latter corresponds to the first term in Eq. (54), it is easily verified that Smit's claim is correct within our simplifying assumptions that τ_q and τ are constant. This fact has also been recognized by Adams and Blount.[3] In their quantum-mechanical calculation, Adams and Blount found, in addition, a third contribution whose physical interpretation was not clear. The corresponding term in the present quasi-classical treatment is the second one in Eq. (54), which is found to be quantitatively equal within the same assumptions as above, and which has been readily interpreted as resulting from the depopulation of the states shifted in energy. It is thus found that, although they are formally analogous as demonstrated by Adams and Blount, the spontaneous "drift" current in a crystal is finite in a steady-state situation, but the "spin current" of a Dirac particle always vanishes in such a situation. The difference obviously comes from the statistical feature present in the crystal problem, which furthermore allows for selective changes of occupancy of the particle states.

B. Skew Scattering

Since the third term in Eq. (46) is nondiagonal in the energy representation of H_p, it induces a supplementary scattering, which we call "skew scattering," in addition to the ordinary scattering associated with the real

[14] J. M. Ziman, in *Electrons and Phonons* (Clarendon Press, Oxford, England, 1960), Chap. 12.

[15] S. Doniach, in *Optical Properties and Electronic Structure of Metals and Alloys*, edited by F. Abeles (North-Holland Publishing Co., Amsterdam, 1966), p. 471.

operator $V(\mathbf{n})$. For energy conserving transitions, the fact that the additional scattering operator is pure imaginary and invariant under space inversion implies that the transport effects, which are odd in the magnetization, only occur in odd order of the total coupling energy. While such odd-order terms do occur in approximations to the scattering by impurities higher than the usual Born approximation, they are forbidden in the case of lattice scattering because of the supplementary requirement that the pseudomomentum of the total system has to be conserved in this case. Thus the lifetime of a particle state is limited by interaction processes including only pairs of emission and absorption of field quanta, so that the scattering probability is even in both the real and imaginary parts of the coupling matrix element. Therefore, lattice waves in general (phonons, spin waves, polarization waves) do not produce odd-order transport effects connected with the third term in Eq. (46) and no odd spontaneous Hall effect arises related to the resistance due to these disturbances. This result is in agreement with an analysis of the phonon-limited transverse conductivity by Leribaux,[16] according to the Kubo formula.

The impurity scattering will now be considered in the high dilution limit which, as shown by Kohn and Luttinger,[17] can be evaluated with the scattering potential of a single impurity at the origin:

$$V(\mathbf{n})=V(n);\quad \mathrm{grad}_n V(\mathbf{n})=(dV/dn)\cdot\mathbf{n}/n. \tag{58}$$

The effective Hamiltonian in Eq. (46) becomes, with $\mathbf{L}=\mathbf{n}\times\hbar\mathbf{k}$,

$$H_{\mathrm{eff}}=H_p-(\tau_q/m^*)(dV/ndn)s\mathbf{u}\cdot\mathbf{L}+V(n). \tag{59}$$

The matrix element of the second term of Eq. (59) for transition between plane-wave states $|\mathbf{k}\rangle$ and $|\mathbf{k}'\rangle$ of H_p is given, since $\mathbf{n}$ is odd under inversion, by

$$\langle\mathbf{k}'|(\tau_q/m^*)s\mathbf{u}\cdot\mathbf{L}|\mathbf{k}\rangle=i\hbar\mathbf{k}\times\int\mathbf{n}[(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}]\times\frac{\tau_q}{m^*}\frac{dV}{ndn}\frac{\sin(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}}{(\mathbf{k}-\mathbf{k}')\cdot\mathbf{n}}d\mathbf{n}. \tag{60}$$

For short-range potentials or small momentum transfers, the element becomes

$$\langle\mathbf{k}'|(\tau_q/m^*)s\mathbf{u}\cdot\mathbf{L}|\mathbf{k}\rangle=is\mathbf{u}\cdot(\mathbf{k}'\times\mathbf{k})\bar{\zeta}, \tag{61}$$

with

$$\bar{\zeta}=\frac{\tau_q\hbar}{m^*}\int V(n)d\mathbf{n}. \tag{62}$$

As this matrix element is pure imaginary, it does not contribute in first order to the scattering integral in Eq. (48) within the usual Born approximation. The next order of approximation to the transition probability[17] is given by

$$\delta Q_{kk'}=\frac{2\pi}{\hbar}\delta\omega_{kk'}\sum_{k''}i\pi\delta\omega_{kk''}\times[\phi_{kk''}\phi_{k''k'}\phi_{k'k}-\phi_{kk'}\phi_{k'k''}\phi_{k''k}], \tag{63}$$

with $\omega_{kk'}=\epsilon_k-\epsilon_{k'}$ and $\phi_{kk'}$ the total matrix element for scattering, i.e., within the approximation in Eq. (61),

$$\phi_{kk'}=V_{kk'}+i\bar{\zeta}s\mathbf{u}\cdot(\mathbf{k}'\times\mathbf{k}). \tag{64}$$

Summing over the intermediary states $\mathbf{k}''$ of density D, we obtain to first order in $\bar{\zeta}$:

$$\delta Q_{kk'}=Q_{kk'}2\pi\bar{\zeta}Ds\mathbf{u}\cdot(\mathbf{k}\times\mathbf{k}'). \tag{65}$$

This additional scattering is obviously directional, since it favors certain relative dispositions of $\mathbf{k}$ and $\mathbf{k}'$ with respect to $\mathbf{u}$, and as in Eq. (55), the correction to the transition probability changes sign under interchange of $\mathbf{k}$ and $\mathbf{k}'$ and therefore does not contribute to the production of entropy. With this correction, the Boltzmann equation becomes

$$\frac{-e\mathbf{E}}{\hbar}\cdot\frac{\partial f^0}{\partial\mathbf{k}}=\frac{\partial f_1}{\partial\mathbf{k}}\cdot\left(\frac{\mathbf{k}}{\tau}+\frac{s\mathbf{u}\times\mathbf{k}}{\tau_s}\right), \tag{66}$$

with

$$\tau_s^{-1}=\pi k^2\bar{\zeta}D\int Q_{kk'}\sin^2\theta\frac{d\mathbf{k}'}{8\pi^3}, \tag{67}$$

if $Q_{kk'}$ depends only on θ as in Eq. (50). Equation (66) has the same structure as the Boltzmann equation in a magnetic field and therefore the current

$$\mathbf{j}=-\tfrac{1}{3}e^2\sum_k\tau v^2\times\frac{\partial f^0}{\partial\epsilon}\frac{\mathbf{E}+(\tau/\tau_s)^2s\mathbf{u}(s\mathbf{u}\cdot\mathbf{E})+(\tau/\tau_s)(\mathbf{E}\times s\mathbf{u})}{1+(\tau/\tau_s)^2} \tag{68}$$

comprises and odd Hall-type effect as well as even magnetoresistive-type effects. The trajectories of the particles can thus also be curved as an average result of successive skew-scattering processes. Since both τ^{-1} and τ_s^{-1} are linear in the scattering probability, which is proportional to the density of scattering centers, we find that the corresponding spontaneous Hall effect has a constant angle θ_s independent of the resistivity and given, if $\bar{\zeta}$ and $V_{kk'}$ are constant, by

$$\tan\theta_s=s\tfrac{4}{3}\pi\frac{\tau_q\epsilon_F}{\hbar}D\int V(n)d\mathbf{n}, \tag{69}$$

with ϵ_F the Fermi energy measured from the band edge and τ_q defined by Eq. (43). Since the latter is proportional to m^*, the product $\tau_q\epsilon_F$ is independent of the effective mass; the sign of $\tan\theta_s$ thus is not related to the sign of the charge carrier, but depends in particular on the sign of the scattering potential $V(n)$. If we finally

[16] H. R. Leribaux, Phys. Rev. **150**, 384 (1966).
[17] W. Kohn and J. M. Luttinger, Phys. Rev. **108**, 590 (1957).

assume that this potential is screened by free electrons of density of states D_f at the Fermi surface, we would obtain, according to the Thomas-Fermi model:

$$\tan\theta_s = s\tfrac{4}{3}\pi(\tau_q\epsilon_F/\hbar)Z(D/D_f)\,, \qquad (70)$$

with Z the effective charge of the scattering center. Equation (70) indicates that the skew-scattering effect can be exceptionally large in transition metals. Indeed, in some of these metals, only part of the electrons participate to the screening, so that the ratio D/D_f may be large enough to give rise to a Hall angle of order unity. This mechanism appears to be the physical basis for the giant spontaneous Hall effect found in Fe,[7] since in this metal, the density of states is dominated by d electrons while the screening is limited to the less numerous s electrons.[18]

V. COMBINED GALVANOMAGNETIC EFFECTS

The combined effects of the magnetic induction and the magnetization fields, assumed both parallel to the direction **u**, can be computed by adding all transverse terms in the right-hand sides of Eqs. (51), (56), and (66). In the effective-mass approximation used here, all longitudinal magnetoresistive effects, i.e., for **u** parallel to the electric field **E**, are found to vanish identically, while for **E** normal to **u**, the total current is given by

$$\mathbf{j} = -\tfrac{1}{3}e^2\sum_k \tau v^2 \frac{\partial f^0}{\partial\epsilon}\Big[\mathbf{E}\Big(1+\frac{s\tau_q}{\tau_B}+\frac{\tau_q}{4\tau_s}\Big) + (\mathbf{E}\times\mathbf{u})\Big(\frac{\tau}{\tau_B}+\frac{s\tau}{\tau_s}-\frac{s\tau_q}{\tau}\Big)\Big]\Big[1+\Big(\frac{\tau}{\tau_B}+\frac{s\tau}{\tau_s}\Big)^2\Big]^{-1}. \qquad (71)$$

Three types of galvanomagnetic effects are thus predicted: an even change of the conductivity along **E** associated with the transverse drift, an odd Hall conductivity formed of nonadditive ordinary and spontaneous parts, and magnetoresistive-type changes related to the net curving of particle trajectories. Additional odd effects would result along the magnetization direction if the polarizability $\langle \mathbf{q}_\tau \rangle$ is not parallel to **u**, as expected if degeneracies exist near the Fermi level.

As far as the Hall effect is concerned, the ordinary and spontaneous components of the Hall resistivity or voltage are additive only in the small-field limit where the denominator of Eq. (71) is unity. Then these components can be separated in the experimental data according to a relation of the type

$$\rho_H = R_0 B + R_s M\,,$$

with ρ_H the observed Hall resistivity and R_0 and R_s the respective Hall coefficients. The validity of the small-field limit is, however, beyond the control of experiment, since the saturation magnetization itself produces a large induction field and may also give rise to a large spontaneous angle. In general, therefore, the denominator of Eq. (71) will contain a strong dependence on the applied magnetic field which can be observed in the transverse magnetoresistance. Of course, such dependence is reflected into the Hall voltage variation with the applied field and may severely complicate the task of separating the ordinary and the spontaneous components.

Finally, we remark that, although a net magnetization is an obvious prerequisite for the existence of spontaneous effects, their magnitude is only remotely related to the intensity of the magnetization itself. If, as was supposed in the early papers, independent groups of electrons contribute to the total magnetic moment and to the electrical conductivity, the magnetization does not appreciably affect the Fermi surface of the conduction electrons, and inclusion of the spin-orbit interaction leads to spontaneous effects which are linear in the magnetization. This conclusion is no longer valid in the currently accepted picture of itinerant electron ferromagnetism in the $3d$ transition metals. In the simplest model of this kind, the bands for up- and down-spin electrons are separated by a splitting energy which itself is closely related to the magnetization. Therefore, any changes in the magnetization, for example with temperature, can cause significant changes in the shape and size of the Fermi surfaces, especially if degeneracies lie close to the Fermi level, and such changes can induce strong nonlinearities and sign reversals in the spontaneous effects.

Many of the transport effects described in this paper have, in fact, been observed in Fe, in which they are the strongest, and they have also been found to some extent in Ni and Co. It appears that these spontaneous effects are related to specific features of the Fermi surfaces and, in a forthcoming paper, we shall use the present analysis to investigate the Fermi surfaces and the mean free path distributions in these ferromagnetic metals.

[18] N. F. Mott, Advan. Phys. 13, 325 (1964).