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Hall coefficient of dilute Cu-Au(Fe) alloys. II. Theory

Philip E. Bloomfield,[†] Randall Caton,[‡] and Myriam P. Sarachik*
City College of the City University of New York, New York, New York 10031

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We study the resistivity ρ and the Hall coefficient R for a polycrystalline metallic host which possesses carriers with a variety of transport properties, particularly an anisotropy over the Fermi surface in the background defect scattering rate. Using the s-d model we derive general expressions for ρ and R when the metallic host contains a dilute concentration of magnetic impurities. Our theory when applied to Cu-Au(Fe) correctly predicts the dependence of ρ and R on temperature, magnetic field, and Fe concentration C. The formulas can be expanded in powers of the concentration and we find that the terms in ρ and R which are linear and quadratic in C are consistent with each other and agree with experimental findings reported in a preceding paper.

I. INTRODUCTION

Numerous theoretical studies[1] have been made on materials containing dilute magnetic impurities which exhibit the Kondo effect. Of the many physical properties of these systems which have been investigated, the Hall effect has received relatively little attention. The Hall coefficient has often been used in materials as a standard test for the existence of carriers having different transport properties such as carriers of different effective masses. In the case of an impurity which has a spin magnetic moment, a magnetic field alters the coupling between the impurity moment and the host conduction electrons.

In the presence of an external field H there exist two kinds of carriers, namely, spin up and spin down (with respect to H); and the relative numbers and scattering rates of each spin species vary as a function of field. The Hall coefficient should therefore be a useful investigative tool.

Theoretical studies on the Hall effect in Kondo systems include those by More,[2] by Béal-Monod and Weiner,[3] and by Fert and Jaoul.[4] The first two works include potential scattering and spin-spin scattering from the magnetic impurity as well as electron-spin and impurity-spin coupling to an external magnetic field; they assume that host electronic lifetimes are infinitely long. The third paper discusses the influence of impurity-spin electron-orbital coupling on the Hall coefficient. All three papers assume that there is only one type of carrier in the conduction band.

The paper of More and a fourth paper by Bloomfield, Hecht, and Sievert[5] (BHS) indicate that the resisitivity depends on the sum of the conduction-electron spin-up and spin-down relaxation times, $\tau_\uparrow$ and $\tau_\downarrow$, while the anomalous part of the Hall coefficient depends on their difference. The BHS paper assumes an infinitely long host electronic lifetime and utilizes a symmetric density of states for single carriers, but does not include potential scattering from the magnetic impurity. This paper points out that the spin-flip process for both spin directions is being frozen out by a depopulation of levels as the magnetic field H increases, since there is a decrease in the populations of both the impurity spins and the conduction spins parallel to the magnetic field, this being the state of higher energy. When an H field is turned on at low temperatures, it couples to the individual spins and hence begins to prevent the spin-flip scattering from taking place; both $\tau_\uparrow$ and $\tau_\downarrow$ increase with H and T and consequently the resistivity decreases with increasing H and T. With no asymmetry in the effective density of states, $\tau_\uparrow$ and $\tau_\downarrow$ increase in precisely the same way, so that $(\tau_\uparrow - \tau_\downarrow) = 0$, and there would be no contribution to the anomalous Hall coefficient. More's calculation includes an asymmetry (due to magnetic impurity potential scattering) which allows one of the spin-relaxation times to increase slightly more rapidly than the other with H.

In the preceding paper[6] (hereafter referred to as Hall Effect I) we report on an experimental investigation of the Hall coefficient in Cu-Au alloys containing dilute concentrations of Fe. The materials studied were polycrystalline and included significant background scattering (even in the absence of the Fe impurities) of the conduction electrons. This condition contrasts with the assumptions of the four papers discussed above. Moreover, the study by Fert and Jaoul assumes a strong external field which suppresses spin-flip scattering, and this does not apply to the experimental conditions of Hall Effect I. In order to interpret our experimental findings, we need to utilize a theory which takes into account the effects of significant host scattering as well as a realistic (not free-electron) band structure. Our examination of existing theories did not reveal any treatment which satisfied our requirements.

It is well known that the Fermi surface (FS) of the noble metals is continuous and multiply connected. The FS parameters (such as relaxation time, velocity, mass, and curvature) vary over the FS and they change continuously as a function of Cu-Au alloying. In this paper we study the influence on the anomalous resistivity and Hall coefficient of an anisotropic host-defect relaxation time[7] as well as the magnetic impurity's charge-contrast potential and spin-spin interaction with the conduction electrons in a magnetic field.

In Sec. II A we derive the general expressions for the resistivity ρ and the Hall coefficient R in the presence of a magnetic field for an anisotropic host containing a magnetic impurity. The quantities ρ and R are expressed in a power series in the concentration C. The coefficients are integrals over the FS and will therefore depend on host properties. In the Appendix we discuss several forms of the conductivity tensor for a polycrystalline specimen. In Sec. II B we break up the integrals over the FS into sums of terms, each of which has a different relaxation time τ. We show that for ρ, the linear term in C must be positive and the quadratic term must be negative. For R we show that a linear term exists, and can be of either sign depending on the relative values of the anisotropic relaxation times, velocities, and curvatures. We also show that for a single isotropic carrier in the presence of background scattering, our formula for R reduces to a form which has no linear term in C. In Sec. III A we discuss the FS symmetries and anisotropies in noble metals and their influence on the host Hall coefficient shown in Fig. 1 of Hall Effect I. In Sec. III B we analyze the FS geometry and present values for the anisotropic relaxation times based on calculations and transport and de Haas-van Alphen measurements in noble-metal binary host materials. Section IV uses this analysis to evaluate the general expressions for the Hall coefficient of Cu-Au alloys containing Fe impurities derived in Sec. II B. We then discuss the experimental data from Hall Effect I in the light of these theoretical considerations.

II. THEORY OF RESISTIVITY AND HALL COEFFICIENT IN METALS CONTAINING DEFECTS AND MAGNETIC IMPURITIES

A. General expressions for ρ and R

We take the external static H field along the positive z axis. Then each electronic spin species makes a separate contribution to the conductivity tensor, $\sigma = \sigma^{\uparrow} + \sigma^{\downarrow}$. Each σ is evaluated by integrating in k space over a constant energy surface, $\epsilon_s(k) = \epsilon_F - s\mu_B B$. Spin up or down corresponds here to $s = +1$ or -1, respectively. Due to the fact that the host samples are polycrystalline and disordered alloys, their bulk relaxation times τ are quite short so that $\omega_c \tau \ll 1$. Here ω_c is the cyclotron frequency, eH/mc. We assume that defect scattering (rather than phonon scattering) is dominant at the low temperatures at which the polycrystalline samples are being studied. The total scattering rate of the conduction carriers is given[8] by the sum of the scattering rates due to the host impurities and foreign impurities

$$1/\tau = 1/\tau_0 + C\alpha_s \; . \tag{1}$$

Our use of a binary host alloy and our restriction to low temperatures essentially eliminates the influence of electron-phonon scattering. However, this latter interaction can be implicitly included by regarding $1/\tau_0$ (the scattering rate in the absence of magnetic impurities) and $C\alpha_s$ as the total scattering rates due to defects and lattice vibrations.

In the following, we neglect the spin dependence of everything but the impurity-induced scattering rate α_s. To terms linear in $\omega_c \tau$, we have for the diagonal and off-diagonal conductivity (times H^{-1})

$$\sigma^s = e^2 (12\pi^2 h)^{-1} \oiint_{\epsilon_s} v \tau \, dS_k$$

and

$$\sigma_H^s = -e^3 (6\pi h^2 c)^{-1} \oiint_{\epsilon_s} v^2 \tau^2 \overline{N} \, dS_k \; , \tag{3}$$

where e is the magnitude of the electronic charge, dS_k is a k-space surface element, v is the velocity at point k of the FS, and $\overline{N}$ is the mean curvature of the FS at point k. For further details, see the Appendix of this paper.

We assume the host defect scattering rate is much greater than that due to the (dilute) magnetic impurities and using Eq. (1) we expand τ in powers of C, the fractional concentration of Fe impurity:

$$\tau = \tau_0 - C\alpha_s \tau_0^2 + C^2 \alpha_s^2 \tau_0^3 + \cdots ,$$

$$\tau^2 = \tau_0^2 - 2C\alpha_s \tau_0^3 + 3C^2 \alpha_s^2 \tau_0^4 + \cdots .$$

The resistivity is computed from $\rho = \sigma^{-1}$. Now we obtain

$$\sigma = \sigma^{\uparrow} + \sigma^{\downarrow} = e^2 (6\pi^2 h)^{-1} \sum_{r=0}^{\infty} (-C)^r \sigma_r \; , \tag{4}$$

where

$$\sigma_0 = \oiint_{\epsilon_F} dS_k\, v\tau_0, \quad \sigma_1 = \oiint_{\epsilon_F} dS_k\, v\tau_0^2 \alpha_+ \,,$$

$$\sigma_2 = \sigma_{2+} + \sigma_{2-}, \quad \sigma_{2\pm} = \oiint_{\epsilon_F} dS_k\, v\tau_0^3 \alpha_\pm^2 \,, \tag{5}$$

and

$$2\alpha_\pm = \alpha_\uparrow \pm \alpha_\downarrow \,, \quad \alpha_\uparrow^2 + \alpha_\downarrow^2 = 2(\alpha_+^2 + \alpha_-^2) \,. \tag{6}$$

In terms of these quantities then, in powers of C to order C^2,

$$\rho = \frac{6\pi^2 h}{e^2\sigma_0}\left[1 + \frac{\sigma_1}{\sigma_0} C - \left(\frac{\sigma_2}{\sigma_0} - \frac{\sigma_1^2}{\sigma_0^2}\right) C^2 + \cdots \right] . \tag{7}$$

For the off-diagonal conductivity we carry out a similar calculation and obtain

$$\sigma_H = \sigma_H^\uparrow + \sigma_H^\downarrow = - e^3 (3\pi h^2 c)^{-1} \sum_{r=0}^{\infty} (r+1)(-C)^r \sigma_{Hr} \,, \tag{8}$$

where

$$\sigma_{H0} = \oiint_{\epsilon_F} dS_k\, \bar{N} v^2 \tau_0^2 ,$$

$$\sigma_{H1} = \oiint_{\epsilon_F} dS_k\, \bar{N} v^2 \tau_0^3 \alpha_+ \,,$$

$$\sigma_{H2} = \oiint_{\epsilon_F} dS_k\, \bar{N} v^2 \tau_0^4 (\alpha_+^2 + \alpha_-^2) \,. \tag{9}$$

The Hall coefficient is defined as $R = \sigma_H/\sigma^2$ so that from Eq. (8) and the square of Eq. (7) we find

$$R = (-12\pi^3 \sigma_{H0}/ec\sigma_0^2)(1 + 2R_1 C + 3R_2 C^2 + \cdots) \,, \tag{10}$$

where

$$R_1 = \frac{\sigma_1}{\sigma_0} - \frac{\sigma_{H1}}{\sigma_{H0}} ,$$

$$R_2 = \frac{\sigma_1^2}{\sigma_0^2} + \frac{\sigma_{H2}}{\sigma_{H0}} - \frac{2}{3}\frac{\sigma_2}{\sigma_0} - \frac{4}{3}\frac{\sigma_1\sigma_{H1}}{\sigma_0\sigma_{H0}} \,. \tag{11}$$

When the phonon scattering rates (due to the vibrations of the host lattice and the magnetic impurity atoms) are not completely negligible (but are still small compared to the host defect scattering), we can further expand the τ's and α's appearing in ρ and R. Because of the nonlinear dependence on the σ_i, the resulting expressions do not obey Matthiesen's rule that the phonon and impurity contributions to the resistivity are simply additive.

B. Discussion of linear and quadratic terms in C

Let us now consider the terms in Eqs. (7) and (10); i.e., the anomalous contributions to ρ and R. The linear term (in C) in the resistivity is explicitly positive, while the quadratic term is easily proved to be negative; i.e., $\sigma_2\sigma_0 - \sigma_1^2 > 0$. To see this we represent the integrals over the FS by weighted sums $\int dS_k \cdots = \sum_i w_i \cdots$. Then we have

$$\begin{aligned}
&\sigma_0\sigma_2 - \sigma_1^2 \\
&\quad = \sigma_0\sigma_{2-} + \frac{1}{2}\sum_{ij} w_i w_j v_i v_j (\tau_{0i}^3 a_{+i}^2 \tau_{0j} + \tau_{0j}^3 a_{+j}^2 \tau_{0i}) \\
&\qquad - \sum_{ij} w_i w_j \tau_{0i}^2 v_i a_{+i} \tau_{0j}^2 v_j \alpha_{+j} \\
&\quad = \sigma_0\sigma_{2-} + \frac{1}{2}\sum_{ij} w_i w_j \tau_{0i}\tau_{0j} v_i v_j (\tau_{0i} a_{+i} - \tau_{0j} a_{+j})^2 > 0.
\end{aligned}$$

Note that the term quadratic in C is a sum of two negative terms and thus can derive from two sources: the first term, $-\sigma_0\sigma_{2-}$, depends on the difference between the scattering rates of the spin-up and spin-down electrons (with respect to H), and the second term depends on the anisotropy over the FS of the scattering rates.

The linear contribution to R may be either positive or negative depending on the characteristics of the conduction electrons. Representing the surface integrals by sums again, we have

$$\begin{aligned}
\sigma_0\sigma_{H0}R_1 = \sigma_{H0}\sigma_1 - \sigma_0\sigma_{H1} = -\frac{1}{2}\sum_{ij} w_i w_j \tau_{0i}\tau_{0j} v_i v_j \\
\times (\tau_{0i}\alpha_{+i} - \tau_{0j}\alpha_{+j})(\tau_{0i} v_i \bar{N}_i - \tau_{0j} v_j \bar{N}_j).
\end{aligned} \tag{12}$$

First we note that for $R_1 \neq 0$, we need an anisotropy over the Fermi surface of both the scattering rate ratio $\tau_0\alpha_+$ and the quantity $\tau_0 v \bar{N}$. In real materials these anisotropies are common. We pause now to consider the isotropic case.

For a spherical FS with *one* effective carrier (free-electron model) we have $\hbar k_F = m_0 v_F$, $\sigma_0 = 4\pi k_F^2 \tau_0 v_F$, etc. Here m_0 is the free-electron mass. Then from Eq. (12), $R_1 = 0$, while from Eqs. (5), (9), and (11), $R_2 = \tau_0^2\alpha_-^2$. This was our original expectation: since $e^2\sigma_0/(6\pi^2 h) = me^2\tau_0/m_0$ with $n = \frac{1}{3}\pi(8m_0\epsilon_F/h^2)^{3/2}$, it follows that

$$\begin{aligned}
R &= -(nec)^{-1}[1 + (C\tau_0\alpha_-)^2 + \cdots] \,, \\
\rho &= (ne^2\tau_0/m_0)^{-1}[1 + C\tau_0\alpha_+ - (C\tau_0\alpha_-)^2 + \cdots] \,.
\end{aligned} \tag{13}$$

That is, the primary anomalous contribution of the magnetic impurities to ρ depends linearly on C and depends on α_+, the *sum* of the spin-up and spin-down scattering rates; and the secondary anomalous contribution to ρ depends on the square of C and α_-, the *difference* between the spin-up and spin-down scattering rates. The linear term is missing entirely in the expression for R, and its leading anomalous term is identical to the secondary one of ρ.

For the case of an anisotropic FS, since the argument of the double sum in Eq. (12) depends on the pairing $(i \neq j)$, it is convenient to define ordered pairs $(ij = k)$ such that $\tau_{0i}\alpha_{+i} < \tau_{0j}\alpha_{+j}$; and then we further define

$$\begin{aligned} w_i w_j \tau_{0i}\tau_{0j} v_i v_j &= W_k , \\ \tau_{0i}\alpha_{+i} - \tau_{0j}\alpha_{+j} &= Y_k , \\ \tau_{0i} v_i \bar{N}_i - \tau_{0j} v_j \bar{N}_j &= X_k \ . \end{aligned} \tag{14}$$

Then we also have

$$\sigma_0 \sigma_{H0} R_1 = -\sum_k W_k Y_k X_k \equiv \sum_k Z_k \ . \tag{15}$$

Note that the quadratic term in ρ can be written in these terms as $\sigma_0\sigma_{2+} - \sigma_1^2 = \sum_k W_k Y_k^2$. Since we have defined k such that $Y_k < 0$, contributions to R_1 are positive if $X_k > 0$. That is, $Z_k > 0$ if (a) both contributions are electronlike ($\bar{N}_i$ and $\bar{N}_j$, both positive) or holelike ($\bar{N}_i$ and $\bar{N}_j$, both negative) and

$$|\bar{N}_i / \bar{N}_j| > \tau_{0j} v_j / (\tau_{0i} v_i); \tag{16}$$

or if (b) there is a holelike ($\bar{N}_j < 0$) and an electronlike ($\bar{N}_i > 0$) contribution. To find the net value of R_1 we must sum all the pair contributions Z_k.

III. BINARY NOBLE-METAL ALLOYS: HOST PROPERTIES

A. Behavior of host Hall coefficient and resistivity: Discussion of experimental data

The Cu-Au system allows complete miscibility for all compositions. The two metals are in mutual solid solution[9]; they are alike with respect to valence and crystal structure; however, they have very dissimilar atomic volumes. For the disordered alloy, Nordheim's concentration rule for the resistivity is supposed to hold:

$$\rho_0 - (\text{const.}) \propto X(1 - X) , \tag{17}$$

where ρ_0 is the resistivity of the host alloy without Fe, and X is the at.% Au in the Cu-Au alloy; and indeed we find that the resistivity of our host samples obeys Eq. (17). Our data[10] in fact obeys the formula

$$\rho_0 \approx [0.004 + 52.8X(1 - X)] \ \mu\Omega\,\text{cm.} \ . \tag{18}$$

Now ρ_0 depends on σ_{xx}^{-1} and R_0 goes as $\sigma_{xy}/\sigma_{xx}^2$. Since σ_{xx} depends linearly on τ_0 while σ_{xy} is proportional to τ_0^2, we expect both these quantities to change with composition according to the change in the average τ_0. Because the formula for R_0 effectively divides out the average τ_0 and v, the measurement of R_0 is a sensitive gauge of the changing lattice constant and relative relaxation times. This leads to the complicated compositional behavior of the Hall coefficient R_0 of the host alloy without Fe shown in Fig. 1 of Hall Effect I. (Note that in Hall Effect I, the host Hall coefficient is referred to as R_h.) From this figure and from our ρ_0 data[10] we indeed find that $(-R_0/\rho_0^2)^{-1/2} \sim (-\sigma_{xy})^{-1/2}$ also obeys Eq. (17). This means that the dominant scattering rates, τ^{-1}, change together and that Eq. (17) is obeyed by the average τ^{-1}. The lattice constant increases as more Au is added, and hence the magnitude of the Hall coefficient is expected to increase (since $R \propto 1/k_F^3$, where k_F is the Fermi momentum). This increasing trend is indicated in Fig. 1 of Hall Effect I. The magnitude of R_0 for pure Cu or Au is less than predicted for one electron per atom in the conduction band. This is attributed to holes or negative-curvature contributions to σ_H. In Ziman's[11] calculation the cancellation due to the negative mass necks[12] yielded an R_0 too low in value as compared with experiment. In order to compensate this, Ziman assumed that τ on the necks (τ_n) was less than τ on the belly (τ_b) of the FS. However, Deaton and Gavenda[13] found from their ultrasonic attenuation measurements on reasonably pure Cu that $\tau_n \approx 8\tau_b$. We note that the mean free paths in their samples were limited by impurity (not phonon) scattering. According to Ziman,[14] in nondilute Cu-Au alloys, the difference in atomic volumes could cause τ_n to be less than τ_b. However, Heine[15] points out that Hurd's[16] Hall-effect data on Ag-Au alloys indicate that over a narrow band about the neck center, $\tau_n \gg \tau_b$ even for 50% concentrations. On the FS of Cu and Au there are significant bulges toward the square X faces of the Brillouin zone (BZ); there are negative curvatures associated with the regions where these peaks join the bulk of the FS (the belly) (see Fig. 1 herein). Thus, these areas (c and d on Fig. 2), as well as those at the neck and the juncture of the neck and the belly (f and g on Fig. 2), provide the regions of the FS which reduce the magnitude of R_0. We propose that the Hall-effect data on the Cu-Au system can be explained by the peaks below X having large positive curvature while the necks at L have large negative curvature and τ's larger than τ_b (due to the non-

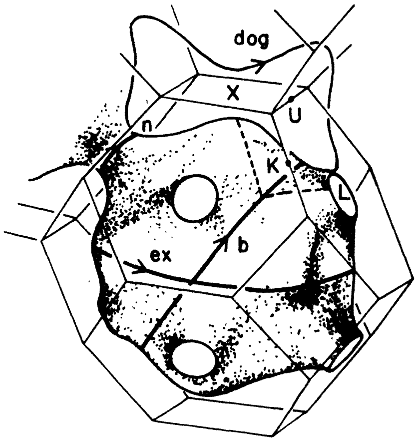

FIG. 1. Brillouin zone and Fermi surface of a noble metal. Four orbits are shown: neck (n), dog's bone (dog), extremal (ex), and belly (b). The basic $\frac{1}{48}$ of the Fermi surface is outlined by the dashed line.

s-state character of the electrons there). Also we require that the τ's from the juncture regions are different from the belly τ. We propose that as the alloy composition changes from pure Cu or pure Au, the τ's from the peaks initially diminish relatively more than the other regions, and hence, these regions contribute somewhat less to R_0. This explains the initial fall of R_0 on either end of Fig. 1 in Hall Effect I. At higher concentrations $(0.25 < X < 0.90) R_0$ rises to larger values, showing the lessening of the negative-curvature contributions due to the relative shortening of τ on the neck regions. (But still we feel the condition $\tau_n > \tau_b$ would hold.)

We wish to comment on the expectations of Ziman[11] that $\tau_n < \tau_b$ in Cu and Cu-Au alloys. Ziman[14] points out that since both Ag and Au have the same atomic volume, the scattering of electrons on the neck (where the electrons are in an admixture of p and d states) would be unaffected by alloying these two metals and the condition $\tau_b \ll \tau_n$ would prevail. For alloys of Cu and Au, however, it is expected that the difference in their atomic volumes would give the same effect as charged impurities and the relative proportions of p-wave to s-wave scattering would increase. We agree with this statement but feel that the relative change would still be small and the condition $\tau_b < \tau_n$ will still hold for Cu-Au alloys as well as for pure Cu or Au. We feel that the reason Ziman had to invoke the condition $\tau_n < \tau_b$ was that in his simplified treatment he only allowed two relaxation times over the whole FS. This contrasts with our differentiating between the τ on the peak, belly, neck, and juncture regions.

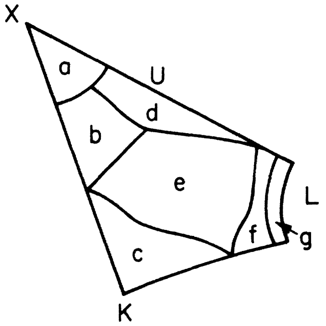

FIG. 2. Basic $\frac{1}{48}$ of the Fermi surface. The points X, U, L, and K correspond to the points on the Fermi surface which are intersected by the line drawn from the center to the matching points on the Brillouin zone. We divide the surface area into the regions a–g.

B. Presentation of approximate anisotropic FS parameters

From an examination of the contour map shown in Fig. 1, from a three-dimensional model of the Cu FS[17] (from which Fig. 1 was drawn), and from FS radii[18] and radii of curvature data[19] taken on Cu, we have taken the basic $\frac{1}{48}$ of the FS (indicated on Fig. 1 by dashed portion) and divided it into seven regions as shown in Fig. 2. We choose radii of curvature to parametrize each region from characteristic points therein. These data are entered in Table I under the row $\overline{N}'$. There are two regions contributing negative mean curvature (f and g encircle each of the necks under $L\langle 111\rangle$). Regions b, c, and d have positive mean curvature, but principal radii of opposite sign. Region a is characterized by the peak under X $\langle 100\rangle$ and has equal positive principal radii. Region e, which we have taken as the most extensive area (see the row w' in Table I), contains both positive and negative principal radii; but the mean curvature is positive everywhere and the value of $\overline{N}'$ in Table I is characteristic of this region (the belly).

The velocities are taken from Halse[18] and Doezema and Koch[19] and are presented in Table I in terms of the free-electron-sphere Fermi velocity, $v_F = 1.578 \times 10^8$ cm/sec. These velocities, as well as the relaxation-time values taken from Atkinson's[20] analysis of his magnetoresistivity measurements, are characterized by typical points in the regions a–g. The true distribution of τ_0 over the FS is probably more anisotropic than Atkinson's analysis indicates. According to Harris,[21] Atkinson used a somewhat simplified parametrization of $\tau(\vec{k})$; however, for our purposes his analysis is sufficient. By identifying the parts of the FS involved in the four orbits measured by Deaton and Gavenda[13] (depicted in

TABLE I. Approximate Fermi surface parameters for copper. The weights, w', are fractions of the basic $\frac{1}{48}$ FS area; $\bar{N}' = 10^8 \bar{N}$ (cm), (Refs. 17–19); $v' = v/(1.578\times 10^8$ cm/sec), (Refs. 18 and 19); $\tau_0' = 7.6\times 10^{14}\, \tau_0$ (sec), (Ref. 20); $1/\tau_s' = (1/\tau_s')_i/(1/\tau_s')_d$, (Refs. 21 and 22).

Region	a	b	c	d	e	f	g
w'	0.092	0.130	0.186	0.115	0.382	0.066	0.031
$\bar{N}'$	$(0.373)^{-1}$ $+(0.373)^{-1}$ $=5.4$	$(0.42)^{-1}$ $+0$ $=2.4$	$(1.30)^{-1}$ $-(1.62)^{-1}$ $=0.16$	$(0.25)^{-1}$ $-(0.68)^{-1}$ $=2.5$	$(1.33)^{-1}$ $+(1.33)^{-1}$ $=1.5$	$(0.31)^{-1}$ $-(0.13)^{1}$ $=-4.3$	$(0.26)^{-1}$ $-(0.10)^{-1}$ $=-6.0$
v'	0.70	0.77	0.70	0.78	0.74	0.60	0.43
τ_0'	1.8	1.7	1.2	2.1	2.7	2.9	4.0
$1/\tau_s'$	0.80	1.40	1.20	1.00	1.12	0.40	0.12
$w'\tau_0' v'$	0.113	0.170	0.156	0.189	0.764	0.115	0.053
$\bar{N}'\tau_0' v'$	6.80	3.14	0.13	4.10	3.40	−7.48	−10.32
$w'\bar{N}'\tau_0'^2 v'^2$	0.77	0.54	0.02	0.77	2.60	−0.86	−0.55
τ_0'/τ_s'	1.44	2.38	1.44	2.10	3.02	1.16	0.48

Fig. 1), we analyzed the relaxation time into four characteristic values (pertaining to the neck, peak, belly, and the region below K). The resulting anisotropy was consistent with Atkinson's relaxation-time map; however, Deaton and Gavenda's data were taken on pure Cu having average $\tau_0 \approx 10^{-10}$ sec while Atkinson's[20] average $\tau_0 \approx 0.4 \times 10^{-13}$ sec. The resistivity of our host polycrystalline samples of pure Cu is comparable with that calculated from a τ approximately equal to that of Deaton and Gavenda's samples [see free-electron calculation in Eq. (13) above]. However, our most studied composition $\mathrm{CuAu}_{14.3}$, for example, has $\rho_0 = 6.4\ \mu\Omega$ cm. which implies, in this case, that $\tau_0 = 0.56\times 10^{-13}$ sec. We note here that from Eqs. (13) and (18) one may use the formula

$$\tau_0 \approx 3.6\times 10^{-13}\ \mathrm{sec}/\rho_0(\mu\Omega\ \mathrm{cm}) . \tag{19}$$

Also note that when we sum the entries in row 8 of Table I, divide by the square of the sum of row 6, and then divide by the corresponding free-electron value $[(\sigma_H/\sigma_0^2)_{\mathrm{fe}} = 2.0]$, we obtain $R_0/R_{\mathrm{fe}} \approx 0.68$. This compares favorably with[11] the observed Cu value, $R_{\mathrm{obs}}/R_{\mathrm{fe}} \approx 0.66$–$0.70$.

We remind the reader that the values of τ presented in the fourth row of Table I are based on relatively pure Cu (Atkinson's samples have $X \leqslant 0.01$). They should all be scaled according to Eq. (19) to give the first-order effect of alloying. Furthermore, as pointed out in Sec. III A, as X (or $1-X$) increases from zero, we expect that τ_a will become slightly reduced relative to τ_b; and then as X takes on values between 0.25 and 0.90, we expect that τ_n will decrease relative to τ_b.

IV. EVALUATION OF THE LINEAR COEFFICIENT IN R FOR Cu-Au CONTAINING DILUTE CONCENTRATIONS OF MAGNETIC IMPURITES

We shall compute the magnetic impurity-induced scattering (α_s) from the papers of BHS[5] and More.[2] These authors take a contact potential for the Kondo scattering, so that their scattering rate is independent of the drift velocity; we make the distinction between the different carriers by writing

$$\alpha_{si} = \alpha_s/\tau_{si}' . \tag{20}$$

Thus the α_s will carry the full temperature and magnetic-field behavior of the scattering rate by the magnetic impurity; while $1/\tau_{si}'$ will describe the dependence on the FS position of the electron's scattering by the Fe.

The FS scattering rate enhancement, $1/\tau_s'(k)$, may be estimated by considering the phase-shift formalism for the total scattering rate in dilute alloys.[21] The host electronic wave functions are expanded in angular momentum components (L) over the FS. The scattering of the host electrons from impurity atoms causes changes in the phases of these components. The contribution to the $1/\tau_s'$ at each point of the FS depends on a sum of components (s, p, or d symmetry) each of which depends essentially on the product of three factors: (i) the amount of s-, p-, or d-like character in the Bloch state (these inverse partial lifetimes are a measure of the s, p, and d charge densities over the FS); (ii) the L-dependent normalization factor for the impurity atom; and (iii) the sine of the phase-shift change due to the impurity atom. Now Coleridge[22] has made an analysis of his de Haas–

van Alphen effect data in dilute Cu(Fe). He has observed the effect of the magnetic-field-dependent exchange scattering (Kondo effect) on the Dingle temperature and has found that the scattering of the neck electrons is unenhanced by exchange effects while the belly electrons suffer increased scattering.

According to Harris,[21] scattering by Fe in noble metals can be described by pure *s*- and *d*-wave terms. The neck region of the Cu FS is predominantly *p* like with some *d*-like character (about 1.8: 1 according to Refs. 21 and 22). Thus, the large charge density of the *p*-wave component of the electrons on the neck is unaffected by the Fe impurities; only the *d*-wave component will participate. However, the Fe contributes both *s* and *d* scattering. The neck electrons have no *s*-wave component, but regions *b*, *c*, *d*, and *e* have the largest *s*- and *d*-wave component; hence the electrons from these latter regions have their (exchange) scattering rate enhanced, while the electrons from the neck region have their scattering diminished. Taking these considerations and the partial lifetime FS maps of Harris[21] into account, we propose the scattering rates $(1/\tau_s')_i$ presented in Table I.

Coleridge[22] notes that the resistivity data on Cu(Fe) can also be understood by assuming that while the belly electrons have their scattering enhanced, the neck electrons are scarcely affected by the exchange scattering from the Fe impurities. It is important for us to point out to the reader (see Harris[21]) that there is an identity between de Haas–van Alphen lifetimes and relaxation times for noble-metal electrons scattering from Fe impurities because of the absence of *p*- wave scattering (the terms contributing to scattering then have the same parity). Next we note that the quantity ρ, taken from Ref. 5 and shown in Fig. 5 of Hall Effect I, is directly proportional to $\alpha_+(\alpha_+ \approx 10^9 \rho N/DC)$. Using this assumption, from the sixth, seventh, and ninth rows of Table I, we can calculate and present in Table II the necessary data for evaluating $\sigma_0 \sigma_{H0} R_1$ in Eq. (15). The sum of the 21 ordered pairings in Table II yields $\sum_k Z_k = \alpha_+ \sum_k Z_k' = -3.73\alpha_+$.

As discussed above, the qualitative effect of alloying is to reduce all the τ's according to Eq. (19). Some of the τ's (such as for regions *a*, *f*, and *g*) may be reduced proportionately more than others; but we expect the relative ordering of the τ's will be maintained. Also, the relative areas and curvatures will change as the alloy composition moves from Cu toward Au; but again we do not expect the qualitative features of Tables I and II to change drastically. Thus the deduction from Table II, that $R_1 < 0$, should hold for the CuAu alloys studied in Hall Effect I; i.e., we expect for most of the materials under our consideration that there is a contribution to the Hall coefficient which is linearly dependent on C and is opposite in sign to the host R_0 [see Eq. (10)]. However, as noted in Hall Effect I, one of the alloy compositions ($X = 0.80$) has a linear contribution of the same sign as R_0 (see Fig. 2 there). We note two other features exhibited by this particular composition: (a) The host R_0 (shown in Hall Effect I, Fig. 1) has a large maximum at the 80% composition. The experimental value has almost reached the free-electron value for R_0. This indicates that the negative contributions to σ_{H0} (f and g in the eighth row of Table I) have decreased while the positive contributions have correspondingly increased. In this case, then, since the host properties are so different from those of other alloy compositions, we might expect that R_1 would be qualitatively different when magnetic impurities are added. (b) Also in Fig. 9 of Hall Effect I, the magnetic-field dependence of R for CuAu_{80} is zero, and, as mentioned in Hall Effect I, there appears to be a sign reversal in $R_{80} - R_{20}$. Of course T_K is very low for the Au-rich alloys so very little H dependence is expected. We see then that even in this anomalous case the magnetic behavior is determined by the transport properties of the host material.

TABLE II. Ordered pairing of Fermi surface regions for evaluating linear contribution to the Hall coefficient R. Here $Y_k' = (\tau_0'/\tau_s')_i - (\tau_0'/\tau_s')_j$, $W_k' = (w'\tau_0' v')_i (w'\tau_0' v')_j$, $X_k' = (\bar{N}\tau_0' v')_i - (\bar{N}\tau_0' v')_j$, $Z_k' = -W_k' X_k' Y_k'$.

k	$-Y_k'$	W_k'	X_k'	Z_k'	k	$-Y_k'$	W_k'	X_k'	Z_k'	k	$-Y_k'$	W_k'	X_k'	Z_k'
ab	0.00	0.0192	3.66	0.066	*db*	0.28	0.0321	0.96	0.009	*ce*	1.58	0.1190	−3.27	−0.614
ac	0.94	0.0176	6.67	0.000	*be*	0.64	0.1300	−0.26	−0.021	*fd*	0.94	0.0217	−11.58	−0.236
ad	0.66	0.0214	2.70	0.038	*fb*	1.22	0.0196	−10.63	−0.254	*fc*	0.28	0.0179	−7.62	−0.038
ae	1.58	0.0863	3.40	0.464	*gb*	1.90	0.0090	−13.46	−0.230	*fe*	0.92	0.0879	−10.88	−0.880
fa	0.28	0.0130	−14.29	−0.052	*cd*	0.66	0.0290	−3.97	−0.076	*gc*	0.96	0.0083	−10.45	−0.083
ga	0.96	0.0060	−17.12	−0.099	*de*	0.92	0.1440	0.70	0.093	*ge*	2.54	0.0404	−13.72	−1.408
cb	0.94	0.0265	−3.01	−0.075	*gd*	1.62	0.0100	−14.42	−0.234	*gf*	0.68	0.0061	−2.84	−0.012

The theory presented here makes definite predictions about the coefficients in the concentration expansion of the expressions for the resistivity and Hall coefficient. The linear term in ρ adds to the host resistivity and has a temperature and field dependence determined by α_+ [see Eqs. (5) and (7)]. As pointed out in Hall Effect I our experimental data follow quite closely this prediction as is shown by the favorable comparison with the calculation of α_+ carried out by BHS. Furthermore, in the analysis of $CuAu_5$ which had a comparatively large impurity concentration, it was found that a small quadratic contribution existed which was opposite in sign to the linear one. This is in agreement with the proof carried out in Sec. II B. There is a contribution to the quadratic term in both ρ and R which depends on α_- and this can be obtained from the calculation carried out by More. However, our measurements are not sufficiently accurate to warrant such a quantitative fit. In Hall Effect I we show that the concentration dependence of R is dominated by the linear term R_1. Therefore, according to Eqs. (5), (9), and (11), the experimental data for $R - R_H$ versus temperature and field should also be in direct correspondence to α_+ calculated in BHS. This is found to be the case; also it is noted that a small C^2 contribution to R exists and the sign of this term is positive for the $CuAu_5$ composition. Unlike the quadratic term in ρ, the expression for R_2 [see Eq. (11)] may be of either sign.

As noted earlier, the sample $CuAu_{14.3}$ was studied most extensively. It obeys[10] the equation

$$\rho = (6.37 + 1450C)\ \mu\Omega\ \text{cm}. \tag{21}$$

Upon comparison with Eq. (7), we see that

$$\sigma_1/\sigma_0 = 1450/6.37 \approx 230. \tag{22}$$

From Fig. 2 of Hall Effect I and from Eqs. (10) and (11) we can obtain

$$-\frac{R-R_0}{2R_0C} = -R_1 = 350 = \frac{\sigma_{H1}}{\sigma_{H0}} - \frac{\sigma_1}{\sigma_0}\ . \tag{23}$$

Thus from Eqs. (22) and (23) we see that σ_1/σ_0 and σ_{H1}/σ_{H0} are of the same order of magnitude. But according to Eqs. (5) and (9), both these quantities have their orders of magnitude determined by the product $\tau_0\alpha_+$. Then from the τ_0 relevant to this composition [see above Eq. (19)] and from the α_+ of BHS (4×10^{15}) we find $\tau_0\alpha_+ \approx 200$. This shows that the anomalous ρ and R are consistent with each other as predicted by our theory; and that their order of magnitude is determined by the host relaxation time and the single-particle exchange scattering rate.

There is a further relation that can be deduced from these observations. We shall show that an explicit form for the H dependence of α_+ may be obtained. Now for low magnetic fields and temperatures, the theoretical expectation[23] is that the anomalous magnetoresistance, $\Delta\rho = \rho(H) - \rho_0$, goes as $-H^2$. From perturbation theory (high-T calculations[24]), $\Delta\rho$ is expected to be directly proportional to $-M^2$, where M is the magnetization due to the magnetic impurities. Schmitt and Jacobs[25] have made a detailed study of the T, C, and H dependence of ρ and $\Delta\rho$ in Cu(Mn). For all studied values of the parameters (dilute and higher concentrations, above and below T_K, and full range of H),

$$\Delta\rho/\rho(0) = -\lambda M^2\ ,$$

$\lambda > 0$ and independent of H. They conclude that this is true under very general conditions.

Moreover, Franken and Van den Berg[26] in their study of the Hall effect in Cu(Mn) and Ag(Mn) have proposed in analogy with ferromagnetism that

$$R = R_0 + R_m M H^{-1}\ ,$$

where R_0 and R_m are independent of H. Hence they deduce that R vs $[\Delta\rho/\rho(0)]^{1/2}H^{-1}$ should yield a straight line; viz.,

$$R = R_0 + R_m \left|\frac{\Delta\rho}{\lambda\rho(0)}\right|^{1/2} H^{-1} = R_0 + R' \left|\frac{\Delta\rho}{\rho(0)}\right|^{1/2} H^{-1}. \tag{24}$$

Their data obey this equation very well; our data (see Fig. 7 of Hall Effect I) also is fit by this relation quite well. Our theoretical and experimental findings for dilute alloys together with Franken and Van den Berg's relation [Eq. (24) here] lead to the result presented in Eq. (27), which is derived as follows.

From Eq. (7) we have

$$\rho = \rho_0\left(1 + \frac{\sigma_1}{\sigma_0}C\right);\quad \rho(0) = \rho_0\left(1 + \frac{\sigma_1(0)}{\sigma_0}C\right);$$

and from Eqs. (5), (9), and (20)

$$\sigma_1 = C_1\alpha_+,\quad \sigma_{H1} = C_2\alpha_+\ ,$$

where C_1 and C_2 are independent of T and H. Thus,

$$\frac{\Delta\rho}{\rho(0)} = \frac{\rho - \rho(0)}{\rho(0)} = [\alpha_+ - \alpha_+(0)]\,CD_1\ , \tag{25}$$

where

$$D_1 = C_1[\sigma_0 + CC_1\alpha_+(0)]^{-1}\ .$$

Similarly from Eq. (24)

$$R - R_0 = 2R_0CD_2\alpha_+, \tag{26}$$

where

$$D_2 = C_1/\sigma_0 - C_2/\sigma_{H0}\ .$$

Now we shall combine Eqs. (25) and (26) which assert that $\Delta\rho/\rho_0$ and R are linearly dependent upon each other, with the nonlinear relation specified by Eq. (24). Equations (25) and (26) inserted into the square of Eq. (24) yield

$$\rho/\rho(0) \approx \alpha_+/\alpha_+(0) = \frac{2}{x^2}[(1+x^2)^{1/2} - 1] , \qquad (27)$$

where

$$x = \frac{4D_2R_0H}{R'} \left(\frac{C\alpha+(0)}{D_1}\right)^{1/2} .$$

This result, Eq. (27), fits quite well the theoretical calculations in BHS and our own data in Hall Effect I; it also yields the correct quadratic dependence on H at low values of the magnetic field, and the observed asymptotic behavior, H^{-1}, at high values.[27] We point out that the high-field log$H$ dependence expected from perturbation theory is only valid over a limited range of field decades, as has also been pointed out by Yosida and Yoshimori.[23]

V. CONCLUSION

A. Concluding statement and comments on skew scattering theory

One of the major observations of our experimental work reported in Hall Effect I is the pronounced linear dependence on the concentration of the anomalous Hall coefficient. Furthermore, the temperature and magnetic field dependence of the linear contributions to the anomalous resistivity and Hall coefficient are almost identical to each other and similar in behavior to the calculation of the resistivity carried out by BHS.[5] Previous theories of the Hall coefficient based on a single-carrier host and a purely s-d interaction did not obtain any linear contribution to R. A linear term is found in the work of Fert and Jaoul;[4] however, this is a perturbational calculation based on the assumption that the H field is sufficiently high to suppress spin-flip scattering. This assumption clearly does not hold for the experimental conditions of Hall Effect I, and the theoretical predictions of Fert and Jaoul are therefore not applicable. After our work was completed, we received a preprint by Giovannini[28] in which he investigates the spin-orbital scattering influence on R. He considers short relaxation times ($\omega_c\tau \ll 1$) but does not include any anisotropy in τ. A linear concentration dependence of R was found to result from the inclusion of a spin-orbit (skew scattering) term in the Hamiltonian, additional to the usual s-d coupling term. The value of the coefficient of the spin-orbital term was deduced by assuming that the experimentally observed linear behavior is due entirely to this term. The spin-orbit term makes no contribution to the term in ρ which is linear in C. We have shown here, however, that from the usual s-d interaction, linear terms in both ρ and R result, the latter existing if anisotropic host background scattering is included, which is the case in most realistic materials. We can use the experimentally determined behavior of ρ to calculate the coefficient of the s-d term in the Hamiltonian. This coefficient can then be used in our theory to calculate the linear behavior of R, which is found to agree with experiment. We find it unnecessary to invoke an additional term to account for the order of magnitude of the observed behavior. An additional skew scattering contribution to R must be either smaller, or at most comparable to the term derived in the present paper. Giovannini[28] suggests that one could extract details of the magnetic impurity state from experimental data on R. We point out that in order to do this, one must separate the two possible linear contributions to R carefully. This might be done by studying the effect of skew scattering on other physical properties.

B. Summary

We have derived general expressions for the Hall coefficient R and the resistivity ρ for a realistic host which has anisotropic relaxation times and contains a dilute concentration of magnetic impurities. Contrary to the expectations based on a single-carrier model, we found that the same linear (in C) terms contribute to R as well as ρ; and this is observed experimentally. We then consider the particular case of Cu-Au(Fe) and calculate the relevant FS parameters from known transport measurements and analyses of noble metals. We have found it necessary to include anisotropy over the FS in the spin-scattering rates as well as the host relaxation times. We find that our theory correctly predicts the dependence of R on temperature, magnetic field, and concentration. Further, our calculation yields results which are the same order of magnitude as the experimental data. In particular, the terms linear in C for ρ and R are numerically consistent with each other; and the small C^2 contributions to ρ and R which we predict are found to be in qualitative agreement with the experimental findings reported in Hall Effect I.

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APPENDIX

Here we derive and discuss the connection between several forms of the diagonal (σ) and off-diagonal (σ_H) components of the conductivity tensor for a polycrystalline specimen. Ziman[11] has presented formulas equivalent to

$$\sigma = e^2(6\pi^2 h)^{-1} \oiint_{\epsilon_F} v\tau \, dS_k \; , \tag{A1}$$

$$\sigma_H = -e^3(12\pi^2 hc)^{-1} \oiint_{\epsilon_F} \sum_{i=xyz} (v^2 - v_i^2) \times M_i^{-1} \tau^2 v^{-1} \, dS_k \; . \tag{A2}$$

The integrals are over the k-space FS ($\epsilon_k = \epsilon_F$) in one Brillouin zone; τ, v, and M^{-1} depend on FS position (i.e., even though evaluated at the same energy, they exhibit anisotropies); and

$$v_i = \hbar^{-1} \frac{\partial \epsilon_k}{\partial k_i} \; , \quad v^2 = \sum_{i=xyz} v_i^2 \; ,$$
$$M_{yx}^{-1} = \frac{\hbar^{-2} \partial^2 \epsilon_k}{\partial k_y \partial k_x} = \hbar^{-1} \frac{\partial v_x}{\partial k_y} \, , \quad M_x^{-1} = M_{xx}^{-1} \, , \cdots \, . \tag{A3}$$

Stern[29] has presented a formula which we generalize to

$$\sigma_H = -e^3(3\pi \hbar^2 c)^{-1} \oiint_{\epsilon_F} v^2 \tau^2 \bar{N} \, dS_k \; , \tag{A4}$$

where $\bar{N}$ is the mean curvature of the FS at point k. Stern took τ as isotropic, and was interested in optical experiments for which case $\omega\tau \gg 1$; hence $-\tau^2 \rightarrow (\omega + i/\tau)^{-2} \approx \omega^{-2}$, and therefore no relaxation time enters into Stern's formula.

From the Boltzmann equation in the relaxation-time approximation,[30] we have for the diagonal and off-diagonal conductivity

$$\sigma_{xx} = e^2(\pi h^2)^{-1} \int_{\epsilon_F} dk_z \oint d\varphi \, m \tau v_x^2 \; ,$$
$$\sigma_{xy} = -\sigma_{yx} = -e^3 H (\pi h^2 c)^{-1} \int_{\epsilon_F} dk_z \oint d\varphi \, \tau v_x \frac{\partial(\tau v_y)}{\partial \varphi} \, . \tag{A5}$$

Here φ, the orbital parameter which runs continuously from 0 to 2π, and the orbital mass m are defined in terms of the k-space electron orbit induced by the magnetic field[31]:

$$m = \hbar (2\pi)^{-1} \oint dk/v_\perp ; \quad d\varphi = (m v_\perp)^{-1} \hbar \, dk ;$$
$$v_x^2 + v_y^2 = v_\perp^2 = v^2 \sin^2\theta \; . \tag{A6}$$

The velocity, v, given in Eq. (A3) is normal to the FS, and $v_\perp$ is the component perpendicular to the z direction (H field). The conductivity for a polycrystalline sample can be symmetrized and averaged over all directions of H. In this case it is convenient to change the integration variables; from Eq. (A6) $dk_z \, d\varphi = \hbar(m v_\perp)^{-1} dk_z \, dk$; then in terms of the local coordinates, dS_k, $dk_z \, dk = (v_\perp / v) dS_k$ and

$$\sigma = \tfrac{1}{2}(\sigma_{xx} + \sigma_{yy}) = e^2(4\pi^2 h)^{-1} \oiint_{\epsilon_F} dS_k \, \tau v_\perp^2 / v \; .$$

Finally, using Eq. (A6) and averaging over all H directions,[32] we obtain Eq. (A1). Now from Eq. (A3) and $v_\perp \partial/\partial k = v_x \partial/\partial k_y - v_y \partial/\partial k_x$, it follows that

$$\sigma_H = \frac{\sigma_{xy}}{H} = \frac{\sigma_{xy} - \sigma_{yx}}{2H} = -e^3(2\pi h^2 c)^{-1} \int_{\epsilon_F} dk_z \oint d\varphi \, \tau^2 \left(\frac{v_x \partial v_y}{\partial \varphi} - \frac{v_y \partial v_x}{\partial \varphi} \right) = -e^3(2\pi h^2 c)^{-1} \oiint_{\epsilon_F} dS_k v_\perp v^{-1} \tau^2$$
$$\times \left(\frac{v_x \partial v_y}{\partial k} - \frac{v_y \partial v_x}{\partial k} \right) = -e^3(4\pi^2 hc)^{-1} \oiint_{\epsilon_F} dS_k \, v^{-1} \tau^2 (v_x^2 M_{yy}^{-1} + v_y^2 M_{xx}^{-1} - 2 v_x v_y M_{xy}^{-1}) \; . \tag{A7}$$

Equation (A7) reduces to Eq. (A2) if we average over all directions of H and take into account the symmetry of the FS [there exist conjugate points such that $v_y \rightarrow -v_y$, but $(2\hbar)^{-1} \partial v_x^2 / \partial k_y = v_x M_{xy}^{-1} \rightarrow +v_x M_{xy}^{-1}$].

A very useful transformation of Eq. (A7) is achieved by defining N,[33,34] the normal curvature of a normal section of the FS. It is equal to the ordinary flexure of the curve formed by the intersection of the FS with the plane formed by $\vec{v}$ (the normal to the FS) and $d\vec{k}$ (the tangent to the orbit). The unit vector along $d\vec{k}$ is given by

$$\hat{k} = (\hat{k}_z \times \vec{v}) / |\hat{k}_z \times \vec{v}| = (-v_y \hat{i} + v_x \hat{j}) / v_\perp \; ,$$

while the rate of change of the normal to the surface with respect to k (along the orbit) is

$$d\hat{v}/dk = d[(v_x \hat{i} + v_y \hat{j} + v_z \hat{k})/v] / dk \; .$$

We have then[33]

$$N \equiv \hat{k} \cdot \frac{d\hat{v}}{dk} = \left(\frac{v_x \partial v_y}{\partial k} - \frac{v_y \partial v_x}{\partial k} \right) \Big/ v v_\perp \; , \tag{A8}$$

so that

$$\sigma_H = -e^3(2\pi h^2 c)^{-1} \oiint_{\epsilon_F} dS_k\, \tau^2 v_\perp^2 N .$$

Note that *dk* on the actual orbit and *dk* in the normal section are equal; N is independent of θ; $v_\perp$ is given by Eq. (A6); and τ depends only on the position on the FS (not the direction of H). After substituting Eq. (A8) into Eq. (A7) and averaging over all orientations of H, we obtain Eq. (A4). Note that the mean normal curvature can be expressed[33] as $\bar{N} = 1/\rho_1 + 1/\rho_2$, where $1/\rho_1$ and $1/\rho_2$ are the principal curvatures of the FS. Now ρ_1 and ρ_2 are the radii of curvature in two mutually perpendicular planes whose intersection is normal to the FS. The combination $1/\rho_1 + 1/\rho_2$ is invariant[35] with respect to rotation of these two mutually perpendicular planes about $\vec{v}$. Also, when the center of curvature of the normal section lies inside the FS, N or ρ is taken as positive; while if the center lies outside the FS, N or ρ is negative [see Eq. (A8)].

We note here that the normal curvature can be calculated in a straightforward fashion from the now standard representation[18] for the FS:

$$C_0 = \sum C_{IJK}[1 - \cos(\tfrac{1}{2} I a k_x) \times \cos(\tfrac{1}{2} J a k_y) \cos(\tfrac{1}{2} K a k_z)] = (\text{const}).$$

Here IJK range over the non-negative integers. This is in the standard form for the parametric definition of a surface. According to Weatherburn,[36] the normal curvature (which is the sum of the principal curvatures at a point on a surface) is an extrinsic property of the surface; and in terms of the surface,

$$(\text{const}) = \phi(x, y, z) ,$$

and

$$K^2 = \left(\frac{\partial \phi}{\partial x}\right)^2 + \left(\frac{\partial \phi}{\partial y}\right)^2 + \left(\frac{\partial \phi}{\partial z}\right)^2 ,$$

we have

$$-\bar{N} = \vec{\nabla} \cdot \hat{n} = \vec{\nabla} \cdot \left[\frac{\vec{\nabla}\phi}{|\vec{\nabla}\phi|} \right] = \frac{\partial}{\partial x} \left(K^{-1} \frac{\partial \phi}{\partial x}\right) + \frac{\partial}{\partial y} \left(K^{-1} \frac{\partial \phi}{\partial y}\right) + \frac{\partial}{\partial z} \left(K^{-1} \frac{\partial \phi}{\partial z}\right) .$$

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†Present Address: National Bureau of Standards, Washington D. C. 20234.

‡Present Address: Department of Physics, University of Illinois, Urbana, Illinois 61801.

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