Magnetoresistance and Hall coefficient of inhomogeneous metals

D. Stroud and F. P. Pan
Department of Physics, The Ohio State University, Columbus, Ohio 43210
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In strong magnetic fields, most metals have highly anisotropic transport coefficients, and these have long been known to be much influenced by sample inhomogeneities. This paper reports a detailed theoretical study of such effects. Various approximations for calculating the effective transport coefficients of inhomogeneous solids are rederived from a unified point of view; these are then used for a variety of model calculations appropriate to metals with open Fermi surfaces. A small concentration of crystallites with open orbits embedded in a free-electron metal is shown to give rise to a strictly linear transverse magnetoresistance (TMR) at strong magnetic fields. The linear coefficient is strongly dependent on the orientation of the open orbit in the plane perpendicular to the magnetic field $\vec{H}$. Extended-orbit crystallites in a free-electron metal produce a TMR which is initially linear, but saturates at sufficiently strong field. The Hall coefficient R_H is unchanged from its free-electron value to first order in the concentration of defects. A striking geometrical effect is predicted, the TMR from open-orbit crystallites saturating in geometries such that current distortions are unable to propagate parallel to $\vec{H}$. The TMR and Hall coefficient of a free-electron metal containing a larger concentration of open-orbit crystallites is calculated in the effective-medium theory (EMT). The TMR is found to saturate at strong fields, in agreement with previous results of Stachowiak, while the Hall coefficient falls off as $1/H^2$ at strong fields, except in flat-plate samples with $\vec{H}$ perpendicular to the plate, in which case it is predicted to saturate at its free-electron value for a sufficiently large concentration of open-orbit crystallites, but to fall off quadratically for lower concentrations. In contrast, calculations within a non-self-consistent approximation give a strictly linear TMR and a Hall coefficient which saturates at a value below the free-electron coefficient. Possible explanations for the discrepancy are discussed. Calculations in the EMT for a model polycrystal with extended-orbit crystallites reveal a broad field region of quasi-linear magnetoresistance, as found previously by Stachowiak, and a reduced Hall coefficient, as well as a conspicuous geometrical effect. The possible relation of these model calculations to experiments of polycrystalline noble metals is examined, but no quantitative theory for these metals is given.

I. INTRODUCTION

According to the symmetry relations of Onsager,[1] the resistance of a metal in an applied transverse magnetic field must be an even function of field strength $\vec{H}$. Thus it is generally expected that the resistivity at high magnetic fields should vary as an integral power of H^2. But this expectation is violated in a number of metals, in which, in a strong field, the resistivity is found to vary roughly as $H^{1.0}$. This behavior occurs in several free-electron metals with a closed Fermi surface, such as K, In, and Al,[2-4] and, in much more conspicuous fashion, in polycrystalline samples of Cu, Ag, and Au.[5] The linear behavior in free-electron metals violates the standard Lifshitz-Azbel-Kaganov (LAK)[6] semiclassical theory of high-field transport, which predicts saturation (resistance approaching a constant value at strong fields) for metals with a closed Fermi surface. This discrepancy has been variously attributed to charge-density waves,[7] Fermi-surface hot spots,[8] and sample inhomogeneities or voids.[9] The linear magnetoresistance seen in the polycrystalline noble metals is less remarkable because of the open Fermi surface that is known to characterize these metals.[10] The open Fermi surface, in single-crystal samples, leads to highly anisotropic resistance in the presence of a strong magnetic field, the resistivity increasing as H^2 for some field directions but saturating for others.[11] The linear increase observed in polycrystalline samples is thought, therefore, to represent some kind of averaged behavior, resulting from the random orientations of the crystallites in a polycrystal.

In this paper, we present a detailed theoretical study of the galvanomagnetic properties of polycrystalline or otherwise inhomogeneous metals. The principal motivation has been to investigate under what circumstances a linear magnetoresistance could result from inhomogeneities. The present work is a sequel to an earlier publication,[12] in which a formula was given for the effect of inhomogeneities in the low-concentration limit. Here we are concerned particularly with inhomogeneities characterized by open Fermi surfaces. We consider such defects both in the low-concentration regime and at larger densities, and treat not only the magnetoresistance but also the high-field Hall coefficient, which also behaves anomalously[13,14] in numerous metals with a linear magnetoresistance. We also show that the results de-

pend strongly on the *shape* of the defect, and, in particular, its orientation relative to the magnetic field.

The essence of the present problem is to calculate the effective conductivity of an inhomogeneous metal, given those of the constituents. In the present context, the conductivities are tensors which may be highly anisotropic in strong magnetic fields. The result is a very complicated situation which has been of necessity treated, in general by simplified approaches. For example, Ziman[15] showed that the experimentally observed transverse magnetoresistance (TMR) and high-field Hall coefficient in polycrystalline Cu could be roughly reproduced by a model in which the Fermi surface of Cu was treated as four cylinders oriented in [111] directions. However, the effective conductivity of the polycrystal was taken simply as the average of the crystallite conductivities. This approximation is valid only when fluctuations in the conductivity are small, a situation that does not prevail in Cu. Shortly after Ziman's work Herring[16] derived the leading term in a perturbation expansion for the effective conductivity tensor in powers of the spatial fluctuations in conductivity. His work demonstrates the possibility of a nonsaturating magnetoresistance even in a metal with a closed Fermi surface, provided the metal is inhomogeneous. However, the results are still limited in validity to small fluctuations. Stroud and Pan[12] obtained exact results at low concentrations but strong scattering (i.e., large conductivity fluctuations), and demonstrated that a small concentration of voids in a free-electron metal would lead to a strictly linear transverse magnetoresistance at strong fields. Similar results were obtained contemporaneously by Sampsell and Garland[17] via direct integration of the power dissipated in the vicinity of a void, and the predictions have, in fact, been experimentally confirmed by implantation of voids in In.[18]

The most extensive previous work involving open Fermi surfaces was carried out by Stachowiak[19-22] using a self-consistent embedding approximation invented by Bruggeman,[23] and later studied by Landauer,[24] who christened it the effective-medium theory (EMT).[25,26] This approach is *not* necessarily limited to small concentrations of defects or small fluctuations, although its exact realm of validity is very difficult to ascertain. Stachowiak found, using this approximation in conjunction with several models for the magnetoconductivity tensor of single crystals, that a quasilinear magnetoresistance could persist over extended ranges of magnetic field strength, but ultimately the resistance deviated from strict linearity. Similar results were found by Dreizin and Dykhne,[27] using qualitative arguments based on a analogies between high-field current flow in metals and convective transport in fluids.

That part of the present work which deals with large concentrations of defects is in many respects an extension of Stachowiak's work. New features include treatment of the high-field Hall coefficient and of various geometrical effects, and the application of a formalism rather simpler and more flexible from a mathematical point of view, than Stachowiak's. We shall also compare the predictions of the EMT with those of a non-self-consistent approach known in optical problems as the Maxwell-Garnett theory (MGT).[28] These turn out to be surprisingly different,[29] and the reason for this will be speculated upon.

We turn now to the body of the paper. Section II presents a brief review, and a new, physically transparent, derivation of the various approximations to be used for the effective conductivity tensor. In Sec. III we give results for small concentrations of open Fermi-surface defects embedded in a free-electron host. Sections IV and V are concerned with model polycrystalline metals at larger concentrations of defects, and Sec. VI is a summary and discussion of the calculations.

II. APPROXIMATIONS FOR THE EFFECTIVE CONDUCTIVITY

The problem discussed here involves calculating the effective conductivity tensor $\vec{\sigma}_{\rm eff}$ of an inhomogeneous metal characterized by a spatially varying tensor $\vec{\sigma}(\vec{x})$. In this section we shall review several approximations for $\vec{\sigma}_{\rm eff}$, each of which is appropriate under different circumstances.

We suppose first that the metal consists of isolated ellipsoidal inclusions of conductivity tensor $\vec{\sigma}_1$ embedded in a background matrix of conductivity $\vec{\sigma}_o$. In the presence of an applied field $\vec{E}_o$, the field inside one of the inclusions can be computed, for a sufficiently low density of inclusions, as if the defect were isolated. The interior field is then uniform and given by[12]

$$\vec{E}_{\rm in} = [\vec{1} - \vec{\Gamma}(\vec{\sigma}_1 - \vec{\sigma}_0)]^{-1} \cdot \vec{E}_0 , \tag{2.1}$$

where $\vec{1}$ is the 3×3 unit tensor and $\vec{\Gamma}$ is an effective "depolarization tensor." The elements of $\vec{\Gamma}$ are

$$\Gamma_{ij} = \oint_{S'} \frac{\partial G(\vec{x}')}{\partial x_i'} n_j \, d^2x' , \tag{2.2}$$

with S' the surface of a defect centered at the origin, n_j a Cartesian component of an outward normal to S', and G a Green's function satisfying

$$\vec{\nabla} \cdot \vec{\sigma}_0 \cdot \vec{\nabla} G(\vec{x} - \vec{x}') = -\delta(\vec{x} - \vec{x}') ,$$
$$G(\vec{x} - \vec{x}') \to 0 \quad \text{as} \quad |\vec{x} - \vec{x}'| \to \infty . \tag{2.3}$$

In like manner the current density within a grain is uniform and satisfies $\vec{J}_{in} = \vec{\sigma}_1 \cdot \vec{E}_{in}$.

The effective conductivity $\vec{\sigma}_{eff}$ may be defined by

$$\langle \vec{J} \rangle \equiv \vec{\sigma}_{eff} \cdot \langle \vec{E} \rangle , \tag{2.4}$$

the angular brackets denoting volume averages. In the situation described above

$$\langle \vec{J} \rangle = \langle \vec{\sigma} \cdot \vec{E} \rangle = \vec{\sigma}_0 \cdot \langle \vec{E} \rangle + f(\vec{\sigma}_1 - \vec{\sigma}_0) \cdot \vec{E}_{in} = \sigma_0 \vec{E}_0 + f\vec{t} \cdot \vec{E}_0 , \tag{2.5}$$

where

$$\vec{t} = (\vec{\sigma}_1 - \vec{\sigma}_0)[\vec{1} - \vec{\Gamma}(\vec{\sigma}_1 - \vec{\sigma}_0)]^{-1} , \tag{2.5a}$$

and f is the volume fraction of the inhomogeneous metal which is made up of defects. In writing Eq. (2.5) we have used the fact that $\langle \vec{E} \rangle = \vec{E}_o$, which follows from the boundary conditions at infinity and which can be proved rigorously by imagining the system to be bounded by a closed surface on which the potential is specified. It follows from Eq. (2.4) that

$$\vec{\sigma}_{eff} = \vec{\sigma}_0 + f\vec{t} , \tag{2.6}$$

a result expected to be valid at low concentrations ($f \ll 1$). Inverting Eq. (2.6) to get $\vec{\rho}_{eff} \equiv \vec{\sigma}_{eff}^{-1}$ and keeping terms up to first order in f gives Eq. (2.7) of Ref. 12.

At larger concentrations of defects, Eq. (2.6) ceases to be valid and one must resort to approximations. Suppose that the inhomogeneous sample is, in fact, present in the form of a polycrystal, such that the ith grain has conductivity tensor $\vec{\sigma}_i$. The effective conductivity tensor of the polycrystal can still be calculated in principle by imagining an external field $\vec{E}_0$ applied to the sample and using definition (2.4). Now, however, the random nature of the polycrystal prevents $\langle \vec{J} \rangle$ from being calculated exactly. We therefore make a *mean-field* assumption[19-26] and calculate the fields and currents inside the ith grain as if the grain were (i) ellipsoidal in shape and (ii) embedded in a uniform medium of conductivity $\vec{\sigma}_{ext}$. Assumption (i) is, of course, not achievable in a polycrystal, since space cannot be filled by a packing of ellipsoids. However, it is still a reasonable hope that the space-averaged fields inside a grain may be given with fair accuracy by the ellipsoidal approximation. Assumption (ii) defines the mean-field nature of the approximation.

With the above assumptions, the fields and currents inside the ith grain are

$$\vec{E}_{in}^{\,i} = [\vec{1} - \vec{\Gamma}_i(\vec{\sigma}_i - \vec{\sigma}_{ext})]^{-1} \cdot \vec{E}_0 ,$$
$$\vec{J}_{in}^{\,i} = \vec{\sigma}_i \cdot \vec{E}_{in}^{\,i} , \tag{2.7}$$

where $\vec{\Gamma}_i$ is given by Eq. (2.2) with the replacement $\vec{\sigma}_0 \to \vec{\sigma}_{ext}$, provided S' is understood to be the surface of the ith grain (assumed centered at the origin).

The effective conductivity is found[26] from Eq. (2.4), using $\langle \vec{E} \rangle = \vec{E}_0$:

$$\vec{\sigma}_{eff} = \vec{\sigma}_{ext} + \langle \delta\vec{\sigma}_i (\vec{1} - \vec{\Gamma}_i \delta\vec{\sigma}_i)^{-1} \rangle \langle (\vec{1} - \vec{\Gamma}_i \delta\vec{\sigma}_i)^{-1} \rangle^{-1} , \tag{2.8}$$

where $\delta\vec{\sigma}_i = \vec{\sigma}_i - \sigma_{ext}$.

Expression (2.8) depends on the choice of $\vec{\sigma}_{ext}$, and this may be guided by the specific geometry of the material under consideration. If the system is a volume fraction f of ellipsoids of conductivity $\vec{\sigma}_1$ embedded in a matrix $\vec{\sigma}_0$, then a reasonable choice is

$$\vec{\sigma}_{ext} = \vec{\sigma}_0 ,$$

which gives

$$\vec{\sigma}_{eff} = \vec{\sigma}_0 + f(\delta\vec{\sigma})(\vec{1} - \vec{\Gamma}\delta\vec{\sigma})^{-1}[1 - f + f(\vec{1} - \vec{\Gamma}\delta\vec{\sigma})^{-1}]^{-1} , \tag{2.9}$$

where $\delta\vec{\sigma} = \vec{\sigma}_1 - \vec{\sigma}_0$. This may be viewed as the matrix generalization of the Maxwell-Garnett approximation for optical properties and has, in fact, been previously derived by other means.[26]

If, on the other hand, the system is a true polycrystal, with no constituent easily interpreted as the host, then an appropriate choice in Eq. (2.8) is the self-consistent one,

$$\vec{\sigma}_{ext} = \vec{\sigma}_{eff} ,$$

which leads to the self-consistency condition

$$\langle \delta\vec{\sigma}_i (\vec{1} - \vec{\Gamma}_i \delta\vec{\sigma}_i)^{-1} \rangle = 0 , \tag{2.10}$$

where $\delta\vec{\sigma}_i = \vec{\sigma}_i - \vec{\sigma}_{eff}$. This is the EMT for anisotropic media, also previously derived in Ref. 26. For spherical inclusions, it reduces to a result found earlier by Stachowiak. Equation (2.10) is in general a matrix equation, and if $\vec{\sigma}_i$ is a function of some continuously varying parameter, such as crystallite orientation in an applied magnetic field, it may be an integral equation.

III. ISOLATED OPEN-ORBIT AND EXTENDED-ORBIT CRYSTALLITES IN FREE-ELECTRON METAL

As a first application of the formalism of Sec. II, we consider a sample of a free-electron metal containing a volume fraction f of anomalous crystallites which have open or extended orbits along some direction in k space. In this case, the effective conductivity tensor is given by Eq. (2.6), with $\vec{t}$ given by Eq. (2.5a). The resistivity tensor, obtained by expanding $\vec{\rho} = \vec{\sigma}^{-1}$ and keeping only terms linear in f, is

$$\vec{\rho} = \vec{\rho}_0 - f\vec{\rho}_0 \vec{t} \vec{\rho}_0 , \tag{3.1}$$

with $\vec{\rho}_0 = \vec{\sigma}_0^{-1}$. The host conductivity tensor $\vec{\sigma}_0 \equiv \vec{\sigma}_{fe}$ has nonzero elements[30]:

$$(\sigma_{fe})_{zz}=\sigma_0\,,$$
$$(\sigma_{fe})_{xx}=(\sigma_{fe})_{yy}=\sigma_0/[1+(\omega_c\tau)^2]\,, \tag{3.2}$$
$$(\sigma_{fe})_{xy}=-(\sigma_{fe})_{yx}=\sigma_0\omega_c\tau/[1+(\omega_c\tau)^2]\,,$$

where σ_0 is the zero-field conductivity, $\omega_c=eH/mc$ is the cyclotron frequency, τ is a characteristic relaxation time, and the magnetic field $\vec{H}$ is taken in the z direction. The conductivity of a defect with an open orbit depends on the orientation of the open orbit in k space, or, equivalently, the direction in which it would carry current in r space. If the open orbit carries current in the x-y plane at an angle ϕ to the x axis, we take the conductivity $\vec{\vec{\sigma}}_1$ of the defect to be

$$\vec{\vec{\sigma}}_1=\vec{\vec{\sigma}}_{fe}+\vec{\vec{R}}^{-1}\vec{\vec{\sigma}}_{00}\vec{\vec{R}},$$
$$(\vec{\vec{\sigma}}_{00})_{ij}=\delta_{ix}\delta_{jx}s_0\{1-1/[1+(\omega_c\tau)^2]\}\,,$$
$$R_{xx}=R_{yy}=\cos\phi\,, \tag{3.3}$$
$$R_{xy}=-R_{yx}=\sin\phi\,,$$
$$R_{zz}=1\,,$$

with the other components of $\vec{\vec{R}}$ vanishing.[30] Here s_0 is a constant, and we are assuming that the open orbit carries current in parallel with the free-electron conductivity. The term $\{\}$ ensures the proper (quadratic) limiting behavior as $\vec{H}\to 0$.

We consider $\vec{\vec{\rho}}_{eff}$ resulting from this model in three geometries: (a) spherical inclusions, (b) cylindrical inclusions with axes in the y direction, perpendicular to the magnetic field, and (c) cylindrical inclusions with axes in the z direction. Figure 1 shows the current flow patterns, magnetic field directions, and crystallite shapes associated with these three geometries. The resulting matrices $\vec{\vec{\Gamma}}$ are readily calculated from definition (2.2) and are found to be as follows:

$$\left.\begin{aligned}\Gamma_{zz}&=-\left(1-\sqrt{1-\epsilon}\,\frac{\sin^{-1}\sqrt{\epsilon}}{\sqrt{\epsilon}}\right)/\epsilon(\sigma_{fe})_{zz}\,,\\ \Gamma_{xx}=\Gamma_{yy}&=\frac{1}{2}\left(1-\frac{1}{\sqrt{1-\epsilon}}\,\frac{\sin^{-1}\sqrt{\epsilon}}{\sqrt{\epsilon}}\right)/\epsilon(\sigma_{fe})_{zz}\,,\end{aligned}\right\} \tag{3.4a}$$
$$\Gamma_{zz}=-(1-\sqrt{1-\epsilon})/\epsilon\sigma_{fe})_{zz}\,,$$
$$\Gamma_{xx}=\left(1-\frac{1}{\sqrt{1-\epsilon}}\right)/\epsilon(\sigma_{fe})_{zz}\,, \tag{3.4b}$$
$$\left.\begin{aligned}\Gamma_{yy}&=0\,,\\ \Gamma_{zz}&=0\,,\\ \Gamma_{xx}=\Gamma_{yy}&=-1/2(\sigma_{fe})_{xx}\,.\end{aligned}\right\} \tag{3.4c}$$

Here $\epsilon=1-(\sigma_{fe})_{xx}/(\sigma_{fe})_{zz}$ is the "anisotropy" of the background medium.

Figure 2 shows the TMR for a sample of a free-electron metal containing a volume fraction $f=0.01$ of open orbits, as calculated from Eq. (3.1). (The TMR is defined as $\Delta\rho_{xx}=[\rho_{xx}(H)-\rho_{xx}(0)]/\rho_{xx}(0)$. The angle ϕ characterizing the direction in the x-y plane is taken to be 45°, and results are shown for all three geometries (a), (b), and (c). The strength s_0 of the open-orbit conduction has been arbitrarily chosen as 0.1. After an initial quadratic rise, the TMR is seen to increase linearly in H for cases (a) and (b), but in case (c) the TMR saturates at a relatively low value of field. The asymptotic behavior is readily found, both analytically and numerically, to be strictly linear in field; the asymptotic slopes are, however, strongly dependent on ϕ. This anisotropy is illustrated in Fig. 3, which shows $\Delta\rho_{xx}(\omega_c\tau)$ for $\omega_c\tau=200$ (corresponding to a field of 10^4–10^5 G in a pure metal at low

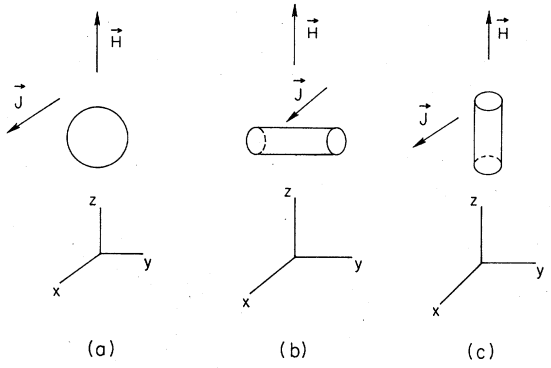

FIG. 1. Crystallite shapes in relation to direction of magnetic field and current flow for calculations described in the text. Configurations (a), (b), and (c) represent a spherical crystallite, a cylindrical one with axis perpendicular to $\vec{H}$, and a cylinder with axis parallel to $\vec{H}$, respectively.

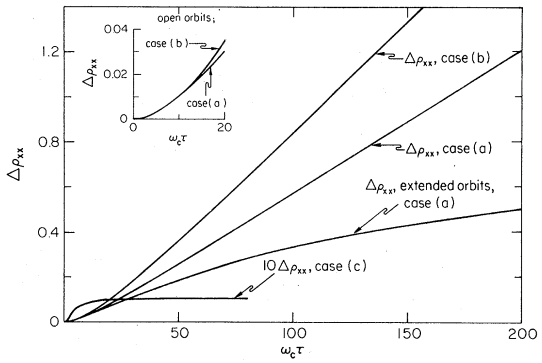

FIG. 2. Transverse magnetoresistance $\Delta\rho_{xx}(H)=[\rho_{xx}(H)-\rho_{xx}(0)]/\rho_{xx}(0)$ for a free-electron metal containing a volume fraction $f=0.01$ of open-orbit crystallites oriented so as to carry current at an angle $\phi=45°$ to the x axis in the x-y plane. Cases (a), (b), and (c) correspond to the geometries shown in Fig. 1. $\omega_c\tau=(eH/mc)\tau$ is a dimensionless measure of magnetic field strength. Also shown is the TMR for extended-orbit defects in geometry (a).

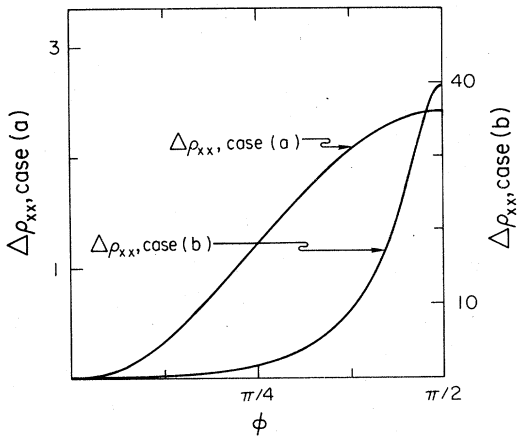

FIG. 3. $\Delta\rho_{xx}(\omega_c\tau=200)$ for a volume fraction $f=0.01$ of open-orbit crystallites embedded in a free-electron metal. The azimuthal angle is described in the text, and configurations (a) and (b) are exhibited in Fig. 1. The remainder of the "rotation" diagram can be obtained from the figure by means of the symmetry relations $\Delta\rho_{xx}(\omega_c\tau,\phi)=\Delta\rho_{xx}(\omega_c\tau,\phi+\pi)=\Delta\rho_{xx}(\omega_c,\tau,\pi-\phi)$.

temperatures) in cases (a) and (b). In both instances, the largest TMR $\Delta\rho_{xx}$ occurs at an angle corresponding to maximum open-orbit conduction in the y direction.

We have also computed the Hall coefficients R_H for the three geometries described above. R_H is defined by the relation

$$R_H=[\rho_{xy}(H)-\rho_{xy}(-H)]/2H\,, \tag{3.5}$$

and for the present case, is found to be identical, through *first* order in f, with the corresponding *free-electron* Hall coefficient defined by

$$R_H^{fe}=[\rho_{xy}^{fe}(H)-\rho_{xy}^{fe}(-H)]/2H\,. \tag{3.5a}$$

Deviations of the ratios R_H/R_H^{fe} from unity occur in *second* order of f, as discussed below.

The linear TMR in cases (a) and (b) are the direct result of current distortion around each anomalous crystallite. As discussed previously by other authors,[12,17] an applied magnetic field is expected to produce, in the vicinity of each anomalous crystallite, a "current shadow" and hence an extra dissipation extending from the defect a distance of order $a(\omega_c\tau)$ parallel to the field, a being a linear dimension of the defect (the radius of the sphere or cylinder). In case (c), the axis of the cylinder is parallel to the defect, the current distortion cannot propagate, and there is no linear TMR.

Equation (3.1) can also be used to calculate $\vec{\vec{\rho}}_{eff}$ and $\Delta\rho_{xx}(H)$ of a sample of a free-electron metal containing *extended* orbit crystallites. An extended orbit is an orbit which spans many Brillouin zones; it is expected to behave like an open orbit for fields sufficiently low that an electron cannot complete a full circuit without being scattered out of the orbit, but it will act like a closed orbit at very high fields.[31] We have computed $\Delta\rho_{xx}$ for extended orbits described by Eq. (3.3) with $\vec{\vec{\sigma}}_{00}$ replaced by $\vec{\vec{\sigma}}_{e0}$ with

$$(\vec{\vec{\sigma}}_{e0})_{ij}=\delta_{ix}\delta_{jx}s_0\left(1-\frac{\tanh x}{x}-\frac{1}{1+(\omega_c\tau)^2}\right), \tag{3.6}$$

where $x=(\pi/2)(dk/\omega_c\tau'\theta)$. This is a model introduced by Stachowiak to describe extended orbits occurring near [111] directions in Cu. The model assumes a "crimped cylindrical" Fermi surface consisting of sections of spheres strung together like beads but attached at the periodic extensions of [111] Brillouin-zone faces. Equation (3.6) results from an evaluation of the Shockley tube integral formula at sufficiently high fields. The model parameters are θ, the angle between the cylinder axis and the plane perpendicular to $\vec{H}$ (the "colatitude"); τ', the relaxation time characterizing electrons on this cylinder; and dk, the ratio of the diameter of the [111] Fermi-surface "neck" in Cu to the Brillouin-zone "diameter", i.e., the distance between opposite hexagonal faces of the Brillouin zone.

Figure 2 shows the TMR in geometry (i) for an extended orbit described by Eq. (3.6). The parameters chosen are $\tau'=\tau$, $\theta=0.045$ rad, $dk=0.18$, and $s_0=0.1$. The value of dk is that which describes the Fermi-surface necks in Cu, but our calculations are not intended to describe polycrystalline Cu but rather to illustrate the general effects to be anticipated from defects with extended orbits. As may be seen from Fig. 2, the TMR associated with the extended orbit does indeed rise linearly with H to about $\omega_c\tau=60$; above this field the slope decreases and at sufficiently high fields (not shown) $\Delta\rho_{xx}$ saturates, as is to be expected of *closed* orbits. The Hall coefficient, as in the open-orbit case, is identical, to first order in f, with the free-electron value of R_H.

IV. POLYCRYSTALLINE METAL WITH OPEN-ORBIT CRYSTALLITES

We turn next to the study of a model polycrystalline metal with an open Fermi surface. The polycrystal is regarded as consisting of two kinds of crystallites. Those of the first kind are oriented in such a way that no open orbits lie in the k_x-k_y plane. These are assumed to have a free-electron conductivity tensor of the form (3.2). The crystallites of the second kind do have open orbits in the k_x-k_y plane which therefore contribute to the conductivity. A crystallite of this kind having an open orbit which makes an angle ϕ to the k_y axis is as-

sumed to have conductivity of the form (3.3). We calculate the effective conductivity for this model within the EMT and, for a particular geometry, also in the MGT.

If f is the volume fraction of open-orbit crystallites, and these point with equal probability in any direction in the x-y plane, the EMT self-consistency condition (2.10) takes the form

$$(1-f)(\vec{\sigma}_{fe}-\vec{\sigma}_{eff})[\vec{1}-\vec{\Gamma}(\vec{\sigma}_{fe}-\vec{\sigma}_{eff})]^{-1} + \frac{f}{2\pi}\int_0^{2\pi} d\phi\,[\vec{\sigma}_1(\phi)-\vec{\sigma}_{eff}]\{\vec{1}-\vec{\Gamma}[\vec{\sigma}_1(\phi)-\vec{\sigma}_{eff}]\}^{-1}=0\,, \tag{4.1}$$

with $\vec{\Gamma}$ defined by Eqs. (2.2) and (2.3), provided $\vec{\sigma}\equiv\vec{\sigma}_{eff}$ in Eq. (2.3). We consider Eq. (4.1) for the three geometries discussed in Sec. III, namely, (a) spherical crystallites, (b) cylinders parallel to the y axis, and (c) cylinders in the z direction ($\vec{H}\parallel\hat{z}$ in all three cases). The three configurations may be thought of as idealizations of sample geometries attainable in a real polycrystal. For example, a thick wire, the diameter of which is much larger than that of a typical crystallite, may be approximated as consisting of spherical crystallites (the grains are certainly spherical on the average), while a flat plate which is thin in comparison with a crystallite diameter can be viewed as a section of a larger sample made up of right circular cylinders perpendicular [case (b)] or parallel [case (c)] to the field. In writing Eq. (4.1), we are, of course, assuming that all crystallites have the same shape (though not necessarily the same size), so that $\vec{\Gamma}$ is the same for all.

For the three sample configurations considered here, Eq. (4.3) must be solved numerically. It is found in cases (a) and (c) that $\vec{\sigma}_{eff}$ has the symmetry of $\vec{\sigma}_{fe}$, so that the expression given in Sec. III for $\vec{\Gamma}$ in these cases applies equally to the present calculations, provided $\vec{\sigma}_{fe}$ is replaced by $\vec{\sigma}_{eff}$ in those expressions. In case (c) $(\vec{\sigma}_{eff})_{xx}$ and $(\sigma_{eff})_{yy}$ are unequal, but expression (3.4c) for $\vec{\Gamma}$ still holds with the replacement $\vec{\sigma}_{fe}\to\vec{\sigma}_{eff}$. In all three cases it is found that

$$(\sigma_{eff})_{zz}=(\sigma_{fe})_{zz}=\sigma_0\,, \tag{4.2a}$$

$$(\sigma_{eff})_{xy}=(\sigma_{fe})_{xy}\,. \tag{4.2b}$$

Result (4.2b) can in fact be shown to hold *exactly* for the model stated, not just within the EMT; this follows from the result of Dreizin and Dykhne[17] that if the antisymmetric part of $\vec{\sigma}$ is position independent and equal to $\vec{\sigma}^a$ then the antisymmetric part of $\vec{\sigma}_{eff}$ also equals $\vec{\sigma}^a$. The remaining elements, $(\sigma_{eff})_{xx}$ and $(\sigma_{eff})_{yy}$, differ from their free-electron values, and can be shown to satisfy complicated algebraic equations in each case, which we have solved numerically.[32]

In Figs. 4 and 5 we plot $\Delta\rho_{xx}$ and R_H/R_H^{fe}, as cal-

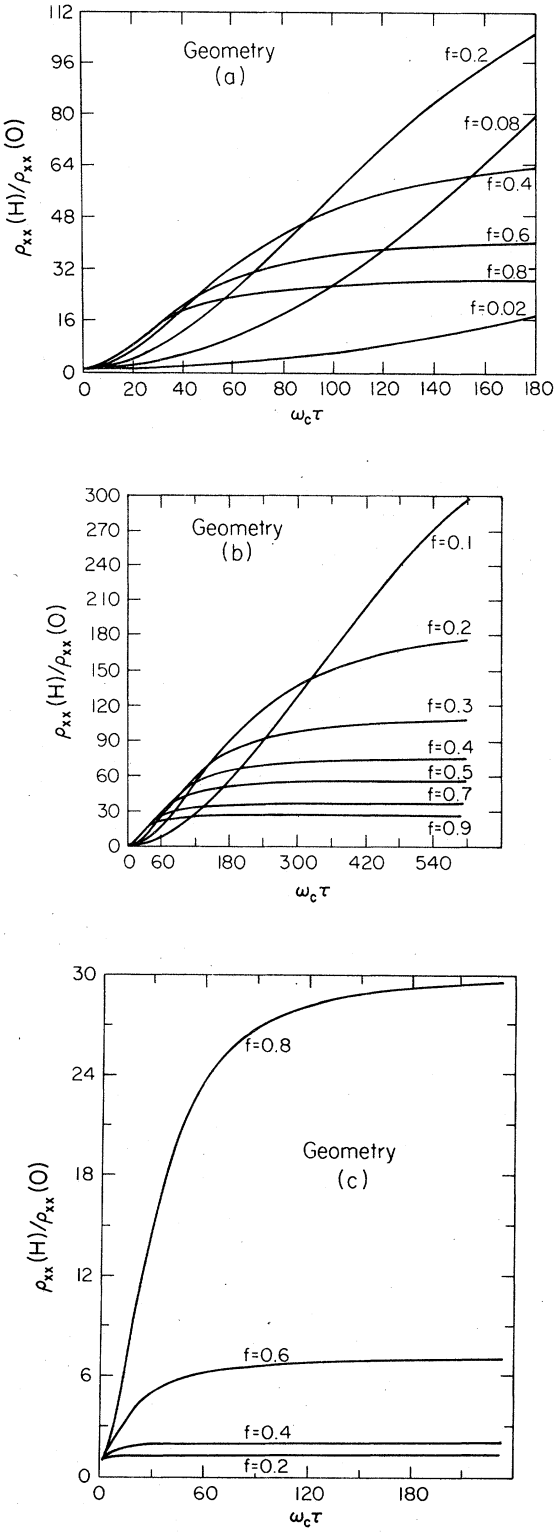

FIG. 4. $\rho_{xx}(H)/\rho_{xx}(0)$ for a volume fraction f of open-orbit crystallites embedded in a free-electron metal, as calculated in the EMT. Configurations (a), (b), and (c) are as in Fig. 1.

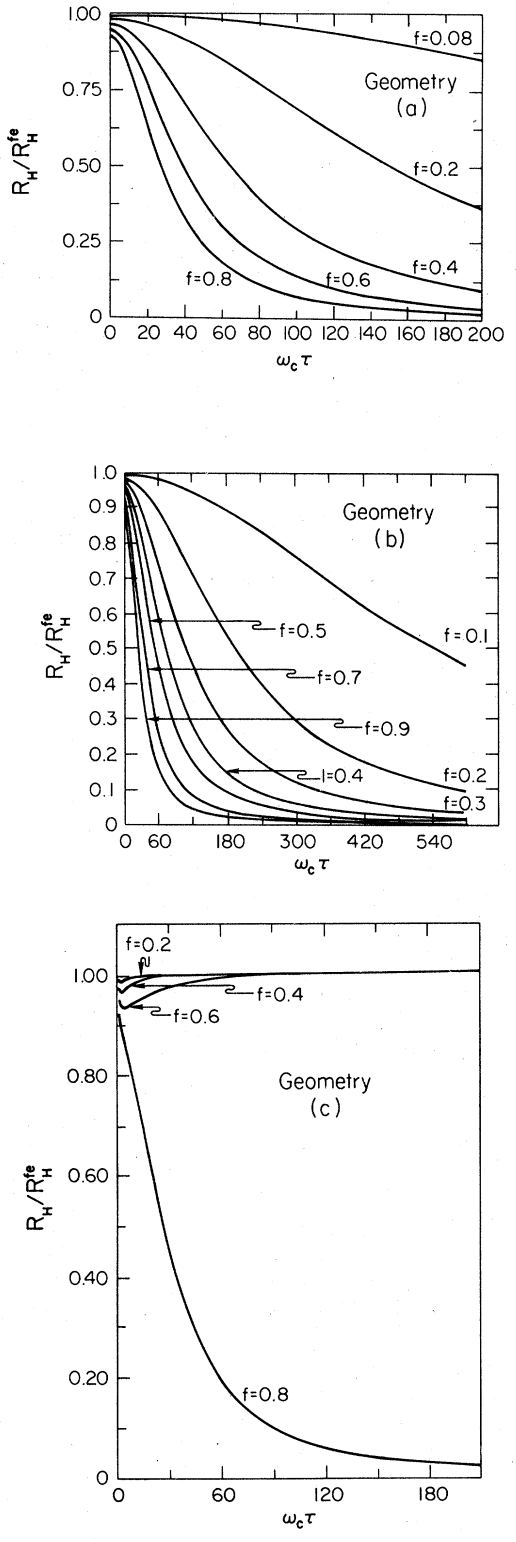

FIG. 5. R_H/R_H^{fe} for a volume fraction f of open-orbit crystallites in a free-electron metal, as calculated within the EMT.

culated from Eq. (4.1), for the three sample configurations and for a variety of values of f. We have arbitrarily chosen $s_0 = 0.1$ in all cases shown.

In geometries (a) and (b) $\Delta\rho_{xx}$ saturates at sufficiently strong fields; the results for ρ_{xx} in case (a) resemble those of Ref. 21, as expected. The deviation of $\Delta\rho_{xx}$ from a strictly linear behavior in these geometries is a result of "interactions" between neighboring open-orbit crystallites, that is, the overlap of their patterns of current distortion. The smaller the value of f, the larger the field at which interaction effects set in and cause saturation. The saturation value of $\Delta\rho_{xx}$ is found to be approximately proportional to $1/f^2$, again in agreement with the qualitative prediction of Refs. 26 and 27.

Although the TMR saturates in this model just as it does in a metal with a closed Fermi surface, the conduction mechanism in the present case is quite different from the latter situation. Here the current in the high-field limit is expected to flow entirely *parallel* to the magnetic field lines, being able to move in a transverse direction only when it encounters an occasional "anomalous" (open-orbit) crystallite. No matter how small the concentration of such crystallites, it appears that the current can always find a transverse path across the sample at sufficiently high fields by this "hopping" mechanism. Thus one expects a field-independent transverse conductivity and, likewise, a field-independent $\Delta\rho_{xx}$, in the high-field limit.

A reflection of this behavior is to be found in R_H corresponding to cases (a) and (b). From Figs. 5(a) and 5(b) it may be seen that R_H falls off as $1/H^2$ at large fields. This stands in contrast to the saturating behavior ($R_H \to$ const) predicted by standard theory[6] for most metals with closed Fermi surfaces. The reason why R_H must go to zero at large fields is clear on physical grounds. If a current density $\vec{J} = J\hat{x}$ is injected into the sample (and $\vec{H} = H\hat{z}$), then HR_H is the voltage difference which must be applied in the y direction in order to maintain the current in the x direction. Because of the peculiar "hopping" method by which the current progresses in the high-field limit, this voltage must asymptotically approach zero, and hence R_H must also.

The situation is completely different in case (c). Here we are dealing with a quasi-two-dimensional pattern of electric fields and current flow. The current cannot flow parallel to the field lines, so the "hopping" mechanism for transverse current flow is not possible. What we have instead, in this somewhat arbitrary model, is essentially a novel kind of percolation conduction[33] in two dimensions. The closed-orbit (i.e., free-electron) crystallites have transverse conductivities which

drop off as $1/H^2$ at large fields. The anomalous (open-orbit) crystallites have field-independent conductivities. The percolation limit is $f=\frac{2}{3}$ in this model (in the EMT), not $f=\frac{1}{2}$ as in more usual two-dimensional (2-D) percolation problems, because the open-orbit crystallites each carry current only in *one* of the two transverse directions, and thus in a sense the effective fraction of anomalous crystallites is less than f, reduced, in fact, by a factor of 2 in the EMT.

It is of interest to compare these results, all found within the same approximation, to analogous results found within the non-self-consistent MGT. We have made such a comparison for geometry (a). If the free-electron metal is regarded as the host, then Eq. (2.8) takes the form

$$\vec{\sigma}_{\rm eff}=\vec{\sigma}_{fe}+f\int_0^{2\pi}\delta\vec{\sigma}_1(\phi)[\vec{1}-\vec{\Gamma}\delta\vec{\sigma}_1(\phi)]^{-1}\,d\phi \times\left(\int_0^{2\pi}[\vec{1}-\vec{\Gamma}\delta\vec{\sigma}_1(\phi)]^{-1}\,d\phi\right)^{-1}, \quad (4.3)$$

with $\delta\vec{\sigma}_1(\phi)=\vec{\sigma}_1(\phi)-\vec{\sigma}_{fe}$. Equation (4.3) is easily solved for $\vec{\sigma}_{\rm eff}$ and the resulting $\Delta\rho_{xx}(H)$ and R_H/R_{fe} are shown in Fig. 6 for $f=0.4$, with other parameters as before. Both curves are remarkably different from the analogous results in the EMT. The magnetoresistance is seen to rise in a strictly linear fashion with magnetic field, just as in the low-concentration regime discussed in Sec. III, and R_H/R_H^{fe} rapidly saturates at a value of ~0.9. This behavior is typical of that we obtain within the MGT at *any* value of f. The asymptotic slope of $\Delta\rho_{xx}$ is found to rise roughly linearly with f; $1-R_H/R_H^{fe}$ rises nearly *quadratically* with f, confirming the low-concentration result of Sec. III that asymptotically $R_H/R_H^{fe}=1$ to first order in f. While it is not claimed that the present calculation is an adequate model for polycrystalline Cu, the results of Fig. 6 are rather similar to those characteristically seen in that metal (the asymptotic Hall coefficient is generally smaller than 0.9 but seems to vary from sample to sample, as discussed below).

The very large discrepancy between the EMT and MGT results is cause for some concern. While the latter seems more in accord with experiment, the EMT agrees better with intuitive pictures of current-flow patterns in a random composite. It is possible that the resolution of the discrepancy lies in the short-range order within the composite. In analogous studies of the optical properties of composites it is found, in materials which consist of metal grains entirely surrounded by dielectric, that the MGT does a superior job of describing the principal absorption peak.[34] This is attributed to the nonrandom nature of the composite.[35] Thus

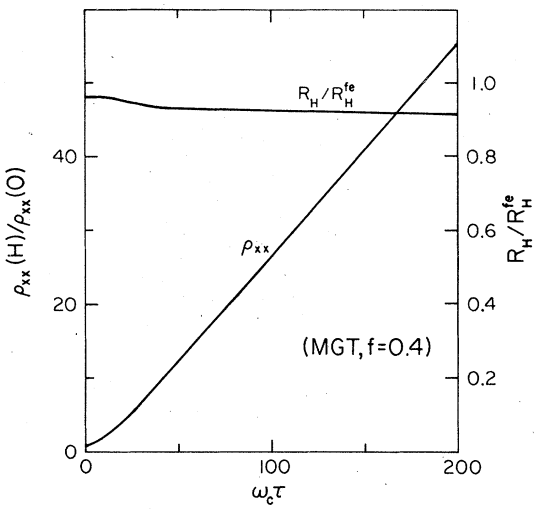

FIG. 6. $\rho_{xx}(H)/\rho_{xx}(0)$ and R_H/R_H^{fe} for a free-electron metal containing a volume fraction $f=0.4$ of open orbits, as calculated within the MGT. Crystallites are assumed spherical [configuration (a)].

one might *speculate* that the MGT, Fig. 6, is better suited to composites in which the open-orbit crystallites are really embedded in a free-electron matrix, while the EMT is most applicable to a more random medium (possibly not achieved in practice).

V. POLYCRYSTALLINE METAL: EXTENDED ORBITS

We next consider briefly a model proposed by Stachowiak to describe the galvanomagnetic properties of noble metals, which takes into account the contributions of extended orbits, as described in Sec. III. In this model, the crystallites are again divided into two classes: (i) free-electron crystallites, with conductivities described by Eq. (3.2), and (ii) extended-orbit grains, with conductivities

$$\vec{\sigma}_1(\theta,\phi)=\vec{\sigma}_{fe}+\vec{R}^{-1}(\phi)\vec{\sigma}_{e0}(\theta)\vec{R}(\phi), \quad (5.1)$$

where $\vec{\sigma}_{e0}(\theta)$ and $\vec{R}$ are defined in Eqs. (3.6) and (3.3) and $\vec{\sigma}_{fe}$ in Eq. (3.2). Crystallites (i) are assumed present in volume fraction $1-f$; those of type (ii) in volume fraction f. If there are four identical cylindrical Fermi-surface sections (as in the noble metals) oriented in [111] directions, and if we neglect the (small) probability that two such cylindrical sections contribute to the conductivity of a given crystallite, then the self-consistency condition (2.10) can be written in the explicit form

$$0=(1-f)\delta\vec{\sigma}_{fe}(\vec{1}-\vec{\Gamma}\delta\vec{\sigma}_{fe})^{-1} +\frac{f}{\Delta\Omega}\int_0^{2\pi}d\phi\int_{-dk}^{dk}d\theta\,\delta\vec{\sigma}_1(\theta,\phi)[\vec{1}-\vec{\Gamma}\delta\vec{\sigma}_1(\theta,\phi)]^{-1}, \quad (5.2)$$

where $\delta\vec{\sigma}_{fe}=\vec{\sigma}_{fe}-\vec{\sigma}_{eff}$, $\delta\vec{\sigma}_1(\theta,\phi)=\vec{\sigma}_1(\theta,\phi)-\vec{\sigma}_{eff}$, and

$$\Delta\Omega=\int_0^{2\pi}d\phi\int_{-dk}^{dk}d\theta\cos\theta\sim 4\pi dk\,,$$

and $f=4\Delta\Omega/4\pi\sim 4dk$ is the probability that one of the four necks will contribute to the conductivity of a randomly oriented crystallite. Equation (5.2) is a matrix integral equation, but the matrix aspect can readily be eliminated and the problem reduced to one of solving a scalar integral equation. We have carried out such a solution for the three geometries discussed previously, using Stachowiak's values $dk=0.18$, which would be appropriate if the model were to apply to the Fermi surface of Cu.[10] We have also arbitrarily taken $s_0=0.27$, and have considered several choices of τ'/τ. The resulting values of $\Delta\rho_{xx}$ are shown in Fig. 7 for geometries (a) and (b). As may be seen, the TMR varies nearly linearly with field in both cases, and depends rather strongly on the ratio τ'/τ. The results in case (a) are nearly identical with those of Stachowiak, a fact which gives us confidence that our formalism reduces to his in the special case of spherical crystallites. In case (c) (cf. Fig. 8), $\Delta\rho_{xx}$ saturates at a very low value of about 4, *irrespective* of τ'/τ. This result indicates that the model is indeed too simple to apply to polycrystalline Cu—the particular oversimplification lying in its neglect of all magnetoresistance in *closed-orbit* directions. In reality, single crystals of Cu generally exhibit a $\Delta\rho_{xx}$ which saturates at a value of at least 10, even in closed-orbit directions,[36] and such an effect would have to be included in a realistic model of polycrystalline noble metals.

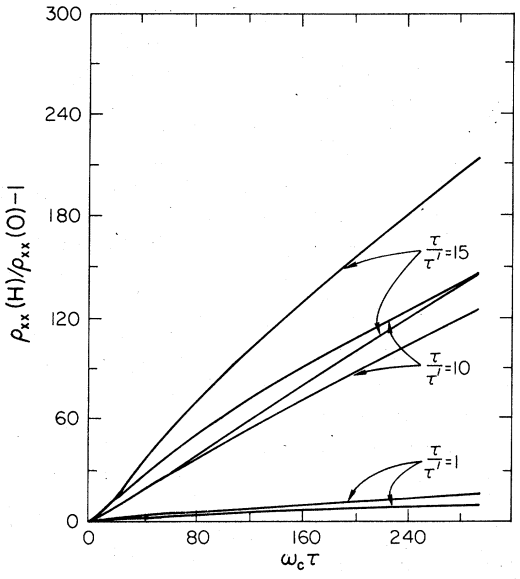

FIG. 7. $\Delta\rho_{xx}(H)=[\rho_{xx}(H)-\rho_{xx}(0)]/\rho_{xx}(0)$ for the polycrystal with extended orbits, as described in the text. The calculations are carried out in the EMT for geometries (a) and (b), as shown in Fig. 1. τ'/τ is the ratio of the relaxation time for small angle scattering to that for scattering from spherical sections of the Fermi surface. For each value of τ/τ', results for geometry (b) lie *above* those for (a).

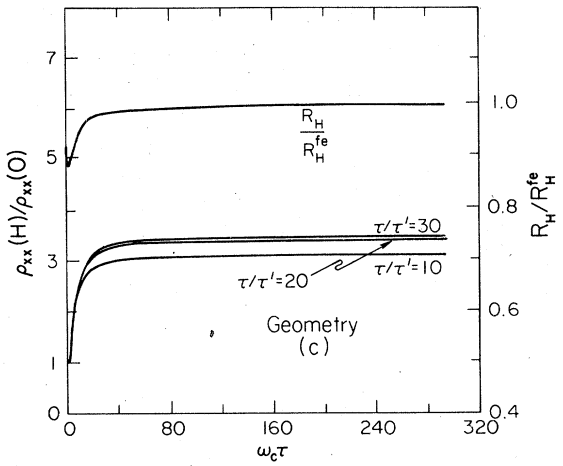

FIG. 8. $\Delta\rho_{xx}(\omega_c\tau)$ and R_H/R_H^{fe} for the extended-orbit model of Sec. V, as calculated within the EMT for geometry (c). The strong field dependence at $\omega_c\tau\lesssim 40$ results from the fact that the model is based on an asymptotic, high-field evaluation of the Shockley tube integral formal for the magnetoconductivity tensor.

The asymptotic behavior of $\Delta\rho_{xx}$ at large fields can also be obtained analytically: we find[32] $\sigma_{yy}\sim H^{-4/3}$ and $\Delta\rho_{xx}\sim H^{2/3}$ in case (a) and $\Delta\rho_{xx}\sim(A+B\ln H)H^{2/3}$ in (b), with A and B constants (the former result in agreement with Ref. 22). The field at which $\Delta\rho_{xx}$ changes from a linear behavior to its asymptotic sublinear form seems to be roughly that at which a substantial fraction of the contributing extended orbits behave as closed orbits. The effect of choosing a large value of τ'/τ is simply to postpone this asymptotic regime, resulting in a larger range of fields in which most of the extended-orbit crystallites behave simply like open-orbit crystallites.

Solution of Eq. (5.2) also gives values of R_H/R_H^{fe}. Results for geometries (a) and (c) are shown in Figs. 8 and 9. [R_H cannot be measured in geometry (b).] At high fields R_H approaches the free-electron value $R_H^{fe}=\sigma_0^{-1}(\omega_c\tau)/H$. At "intermediate" fields ($40\lesssim\omega_c\tau\lesssim 200$) in geometry (a) R_H is slowly varying and smaller than R_H^{fe}. At "low" fields ($\omega_c\tau\lesssim 30$) R_H exhibits spurious behavior resulting from the oversimplified nature of the model, which is based on high-field evaluation of the Shockley tube integral formula. In geometry (c), R_H is field independent for $\omega_c\tau\gtrsim 40$ and nearly equal to R_H^{fe}.

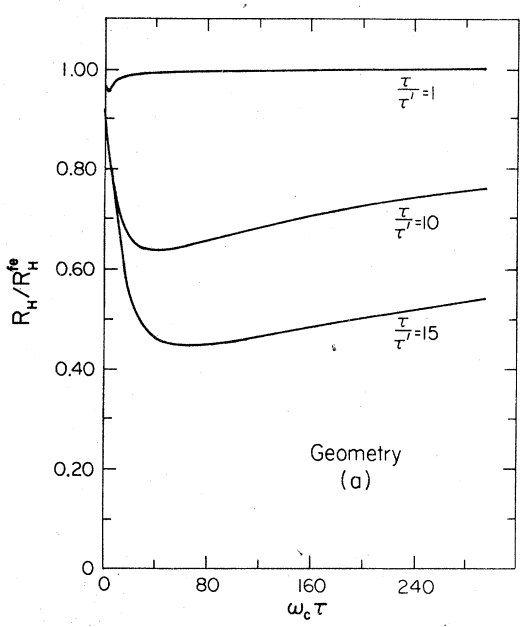

FIG. 9. R_H/R_H^{fe} for the extended-orbit model of Sec. V for geometry (a), as calculated in the EMT. The low-field results ($\omega_c\tau \lesssim 40$) are anomalous for the same reasons as in Fig. 8.

We have not carried out calculations on the Stachowiak model within the *non*-self-consistent embedding approximation. However, the limiting high-field behavior is easy to derive, and we have found, in case (a), that $\Delta\rho_{xx}$ varies asymptotically as $H^{1/2}$ within the MGT, a behavior not very different from that predicted within the EMT.

VI. DISCUSSION

We can draw several conclusions from the results of the preceding sections:

(i) A small concentration of crystallites with open orbits gives rise to a strictly linear TMR in a free-electron metal.

(ii) The TMR is anisotropic, the linear coefficient being strongly dependent on the orientation of the open orbit in the plane perpendicular to the magnetic field.

(iii) There is a striking geometrical effect, the TMR rising linearly only in geometries such that the current distortion produced by a defect can propagate parallel to the field. These are geometries (a) and (b) in the present work.

(iv) At higher defect concentrations, the strictly linear behavior is modified within the EMT, and the TMR ultimately saturates. In contrast, the MGT predicts a continuation of linear behavior. The present work does not permit one to decide which of the two approximations is the more accurate; this may vary from case to case, depending on sample geometry.

(v) In a model sample with extended-orbit crystallites, in geometries (a) or (b), the EMT predicts a quasilinear increase in the TMR over a broad range of fields. Once more, saturation is predicted in geometry (c).

(vi) R_H is found to be affected by open- or extended-orbit crystallites in second order in their concentration. The effect is universally to reduce R_H below its free-electron value, but the manner in which the reduction occurs depends on the approximation used to calculate $\bar{\sigma}_{\text{eff}}$.

It is of interest finally, to consider the relation between these results and experiments in polycrystalline noble metals. These show a linear magnetoresistance in geometries where current distortions can propagate parallel to the magnetic field, saturation in other geometries. The linear rise is here found to be an *exact* property of free-electron metals with a *small* concentration of open orbits, or of crystals with a larger concentration of open orbits, within the MGT. In order to explain the data in Cu,[14] one would require ~40% open-orbit crystallites. We may tentatively conclude that the linear behavior observed in Cu and other noble metals is due to open-orbit defects. The interaction effects predicted by the EMT are not observed, perhaps because the open-orbit crystallites in Cu are for some reason constrained to be separated from one another (short-range order). As far as the Hall data[13,14] on Cu are concerned, most experiments show a field-independent R_H at strong magnetic fields, in agreement with the MGT for open orbits but not the EMT either for open or extended orbits. This result suggests, once again, that perhaps Cu is better described by the MGT than the EMT, for unknown reasons.

ACKNOWLEDGMENTS

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