Conductivity of metallic films and multilayers

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We examine the quantum and semiclassical approaches to the calculation of the electrical conductivity using a model of free electrons with a finite lifetime. We evaluate the Kubo formula exactly for the free-electron model for homogeneous conductors, thin films, and multilayers. We use these results to explicitly demonstrate the relationships among the exact quantum approach, semiclassical approaches, and an approximate quantum approach. One popular semiclassical approach uses a specularity parameter to describe the fraction of electrons that scatter coherently at boundaries. We show that this parameter should depend on the angle at which the electrons are incident on the boundary. Another semiclassical theory employs the concept of a local mean free path. We show that this approximation works surprisingly well; the error in the calculated conductivity being significant only for very thin layers. A third approach uses an approximate solution to the Kubo formula. We show that this approximation works well for thick layers and extremely thin layers but not for intermediate layer thicknesses. We discuss the implication of these results to the study of the giant magnetoresistance.

I. INTRODUCTION

The discovery of the giant-magnetoresistance (GMR) effect[1,2] has renewed interest in the theory of transport in layered and granular systems. Several approaches to transport in layered systems are currently being used. One approach[3–6] is based on the Fuchs-Sondheimer[7,8] extension of semiclassical Boltzmann theory, another[9] is based on the kinetic equation approach of Chambers and Pippard,[10] while a third[11] is based on an approximate solution of the Kubo-Greenwood[12,13] linear response theory. Camblong and Levy[14,15] have recently obtained yet another semiclassical theory for multilayers starting from the Kubo-Greenwood approach.

In this paper, we show that for the model of nearly free electrons with random point scatterers, either in infinite space or in a film of finite thickness, the conductivity can be evaluated rigorously using the Kubo formula and that these results can be used to test and compare all of the proposed theories in considerable detail. Specifically, we show that the semiclassical approaches of Fuchs and Sondheimer, of Chambers and Pippard, and of Camblong and Levy are equivalent for homogeneous systems. For inhomogeneous layered systems, we show that the theory of Camblong and Levy (which we show to be the same as Fuchs-Sondheimer theory with interfacial specularity parameter, p, set to unity) gives conductivities in very good agreement with quantum results obtained from evaluation of the Kubo formula. We also show that the approximate quantum solution of Zhang, Levy, and Fert (ZLF) (Ref. 11) works rather well if all layers are thinner than the shortest mean free path or all layers are thicker than the longest mean free path, but the approximation is rather severe if the layer thicknesses are comparable to the mean free path.

Almost all of the previous work on transport in films and multilayers has been based on the model of free electrons with random point scatterers (FERPS). The justification of the use of the FERPS model to describe the electrical resistivity has been discussed previously.[11,15] It requires one to imagine that the scatterers are points, that they are distributed randomly in space, and that there are many scatterers within a region of space of volume ℓ^3, where ℓ is the mean free path. The assumption of point scatterers is important because such scatterers scatter isotropically. This eliminates the vertex corrections in the linear response formalism or, equivalently, the scattering-in term of the Boltzmann equation approach and thereby allows the use of a relaxation time approximation.[18] Previous experience with silver-palladium alloys[16,17] indicates that the relaxation time approximation is inaccurate for real materials, which have free-electron-like dispersion, because real impurities are not point scatterers. The relaxation time approximation may be reasonable, however, when the Fermi energy falls in the d bands so that the scattering is primarily $s \rightarrow d$ or $d \rightarrow d$, because in this case the vertex corrections tend to vanish because of symmetry considerations.

The assumption of random point scatterers offers an additional simplification. It causes the electron self-energy to be local, which makes it easier to speak in terms of a local scattering rate or a local mean free path.[18] The FERPS model is certainly a major oversimplification, but it has tradition on its side, and it is sufficiently simple that one may hope to see general features without getting lost in detail. It is also useful for testing first-principles calculations, which should be capable of giving the FERPS results in the appropriate limit.

A brief outline of the paper may be helpful to the reader. In Sec. II, we briefly outline the Fuchs-Sondheimer, Chambers-Pippard, Camblong-Levy, and Zhang-Levy-Fert approaches to transport in films and

multilayers. In Sec. III, we calculate the exact nonlocal conductivity using the Kubo-Greenwood formula within the free-electron model for a homogeneous system. In Sec. IV, we take the semiclassical limit of this result and show that it is identical to those of the semiclassical theories of Camblong-Levy, Chambers-Pippard, and Fuchs-Sondheimer. In order to do this, we generalize Fuchs-Sondheimer theory so that it yields the nonlocal conductivity and show that the results are identical to the form proposed by Camblong and Levy. In Sec. V, we present an exact expression for the conductivity of a thin film within the FERPS model. We show the connections between the continuum limit of this expression and the semiclassical theories. We also show that a small change in the semiclassical theory effected by including the zero-point motion of the electrons perpendicular to the film yields an expression that provides accurate fits to experimental thin film conductivities. In Sec. VI, we calculate the conductivity and giant magnetoresistance for several multilayer systems and compare the results to those obtained from the semiclassical theories and the approximate quantum theory of ZLF. We show that for multilayers, the Camblong-Levy theory is equivalent to the Fuchs-Sondheimer theory (with specularity parameter, $p = 1$), while the Chambers-Pippard theory is, in principle, different and more general. We show that Camblong-Levy theory agrees well with the exact solution of the Kubo formula for most layer thicknesses.

II. EXISTING THEORIES OF TRANSPORT IN FILMS AND MULTILAYERS

In this section we very briefly outline the Fuchs-Sondheimer, Chambers-Pippard, Camblong-Levy, and Zhang-Levy-Fert approaches to the electrical conductivity of films and multilayers. This section is not intended as a review of any of these theories. It is included in order to state precisely what the current authors mean by the appellations, Fuchs-Sondheimer, Chambers-Pippard, etc., and to provide the basic equations which we shall need in later sections.

A. Fuchs-Sondheimer theory

Fuchs-Sondheimer theory[7,8] is based on the semiclassical Boltzmann transport equation (BTE) which assumes that one can describe the electrons by a distribution function, $f(\mathbf{p},\mathbf{r},t)$, which gives the number of electrons with momentum $\mathbf{p}$ at point $\mathbf{r}$ and time t. The distribution function is determined by balancing the contributions to the time rate of change of $f(\mathbf{p},\mathbf{r},t)$ under steady-state conditions,

$$\frac{df}{dt} = \frac{\partial f}{\partial t} - \dot{\mathbf{r}}\cdot\nabla f - \dot{\mathbf{p}}\cdot\nabla_p f = 0. \tag{2.1}$$

Considering the departure of f from equilibrium $g(\mathbf{p},\mathbf{r},t) = f(\mathbf{p},\mathbf{r},t) - f_0(E)$, and assuming that the collisions which return the electrons to equilibrium do so in the following simple manner known as the lifetime approximation:

$$\frac{\partial f}{\partial t} = \frac{\partial g}{\partial t} = -\frac{g}{\tau}, \tag{2.2}$$

we have

$$\left[\mathbf{v}\cdot\nabla + \frac{1}{\tau}\right] g(\mathbf{v},\mathbf{r}) = \frac{e}{m}\boldsymbol{\mathcal{E}}\cdot\nabla_{\mathbf{v}} f_0 = e\boldsymbol{\mathcal{E}}\cdot\mathbf{v}\frac{\partial f_0}{\partial E}, \tag{2.3}$$

where we have used $\dot{\mathbf{p}} = -e\boldsymbol{\mathcal{E}}$ and have eliminated $\mathbf{p}$ in favor of the electron velocity $\mathbf{v} = \mathbf{p}/m$. In an application to a real (periodic) system, the velocity would be related to the momentum through the dispersion relation, $v_{\mathbf{k}} = \hbar^{-1}\nabla_{\mathbf{k}} E_{\mathbf{k}}$, where $\mathbf{k} = \mathbf{p}/\hbar$. Once the distribution is determined from solving Eq. (2.3) the current can be calculated from

$$\mathbf{J}(\mathbf{r}) = \frac{-e}{(2\pi)^3}\int d^3\mathbf{k}\ \mathbf{v}_{\mathbf{k}} g(\mathbf{v}_{\mathbf{k}},\mathbf{r}). \tag{2.4}$$

Films and multilayers are treated within Fuchs-Sondheimer theory by assuming different distribution functions, $g^I(\mathbf{v},z)$, within each layer and matching boundary conditions at each interface. Thus, the general solution to Eq. (2.3) within layer I has the form

$$g^I(\mathbf{v},z) = \frac{e\tau_I}{m}\boldsymbol{\mathcal{E}}\cdot\mathbf{v}\frac{\partial f}{\partial E}[1 + F^I(\mathbf{v})e^{-z/\tau v_z}], \tag{2.5}$$

where the functions $F^I(\mathbf{v})$ are to be determined by a matching procedure at the interfaces between layer I and layers $I-1$ and $I+1$. The original application of this approach by Fuchs was to films. He reasoned that space could be divided into three regions, region 1 to the left of the film ($z < 0$), region 2, the film itself ($0 < z < d$), and region 3 to the right of the film ($z > d$). On the left side of the film (at $z = 0$), the distribution function for the electrons should have no electrons with $v_z > 0$ other than those specularly reflected off of the interface because there are no electrons outside the film. Thus supposing that a fraction p_L of the electrons which strike the left boundary is specularly reflected, we must have

$$g_+(\mathbf{v}_+,0) = p_L\, g_-(\mathbf{v}_-,0) \tag{2.6}$$

and similarly at the right side of the film the electrons with $v_z < 0$ could only be those specularly reflected off the boundary,

$$g_-(\mathbf{v}_-,0) = p_R\, g_+(\mathbf{v}_+,0). \tag{2.7}$$

The case of $p_L = p_R = 1$ corresponds to perfect reflection and can be shown to lead to a conductivity for the film identical to that for an infinite homogeneous system. The case of $p_L = p_R = 0$ corresponds to perfectly diffuse scattering, i.e., all electrons which strike the surface are assumed to lose all memory of their velocity before scattering.

The generalization of this theory to interfaces[3–5] is straightforward. Between layers I and $I+1$, it is assumed that a fraction T of the electrons will be transmitted coherently and a fraction R will be reflected coherently. The remainder, $1 - R - T$, are assumed to scatter incoherently. Thus, at the interface between layers I and $I+1$,

$$g_-^i = T^{I,I+1} g_-^{I+1} + R^{I,I+1} g_+^I. \quad (2.8)$$

In general, within the FERPS model, if the real part of the potential changes between layers, the electrons will be refracted at the interfaces,[5] i.e., T and R will depend on the angle of incidence. In this paper, we shall generally assume that the real part of the potential does not change between layers. Although this assumption does not simplify the exact calculations which we present other than by reducing the number of arbitrary parameters, it does make it easier to compare with the Camblong-Levy and Zhang-Levy-Fert theories.

The picture used by Hood and Falicov[5] to relate the FERPS model to layered transition metals puts great emphasis on these steps because it assumes a common band for all electrons, e.g., nickel is treated as a free-electron system with ten electrons per atom. In this picture, layers consisting of different elements will have different numbers of electrons and hence different values of E_F and k_F in the FERPS model. We consider such a picture to be less realistic than the extreme Mott *s-d* picture, which appears to be in the minds of the other authors attacking the GMR problem. In the Mott *s-d* picture, there is a single free-electron band for all of the layers (representing the *s* electrons) and differences in scattering rates between the layers are due, in part, to differing numbers of *d* electrons at the Fermi energy for the different layers.

Ignoring any real potential steps between layers, the boundary conditions at the interface between layers I and $I+1$ become

$$g_-^I = p^{I,I+1} g_-^{I+1},$$
$$p^{I+1,I} g_+^I = g_+^{I+1}. \quad (2.9)$$

We shall show in Sec. V that the specularity coefficients p should depend on the angle of incidence of the electrons on the interface. Note that this is quite different from the angle of incidence dependence of T and R obtained by Hood and Falicov, which originates from the potential steps at the interfaces.

B. Chambers-Pippard theory

Chambers[10] developed the theory of transport from the point of view of kinetic theory. He argued that the energy of an electron passing through point $\mathbf{r}_0$ at time t_0 had had its energy modified by ΔE, where

$$\Delta E(\mathbf{r}_0, t_0) = \int_{-\infty}^{t_0} \mathbf{v} \cdot (-e\boldsymbol{\mathcal{E}}(\mathbf{r}, t)) e^{(t-t_0)/\tau} dt. \quad (2.10)$$

Assuming that the electrons were initially in equilibrium before their energy was modified by the applied field, we have

$$f(\mathbf{r}_0, \mathbf{v}, t_0) = f_0(E) + \frac{\partial f_0}{\partial E} \Delta E(\mathbf{r}_0, t_0). \quad (2.11)$$

This result is, in fact, a solution of the semiclassical BTE. The Chambers-Pippard approach appears to be most useful when the applied field varies spatially or when magnetic fields are present.

If the electron velocity and the lifetime are constant Eq. (2.10) can be written as

$$\Delta E(\mathbf{r}_0, t_0) = -e \int_{-\infty}^{\mathbf{r}_0} \frac{\mathbf{r} \cdot \boldsymbol{\mathcal{E}}(\mathbf{r}, t)}{r} e^{-|\mathbf{r}-\mathbf{r}_0|/\ell} dr, \quad (2.12)$$

where the mean free path is $\ell = v\tau$.

C. Kubo-Greenwood linear response formula

Theories based on the semiclassical Boltzmann transport equation encounter serious conceptual difficulties in dealing with length scales comparable to the electron wavelength. However, fully quantum mechanical expressions for transport coefficients have been derived from linear response theory. Consider a system of noninteracting electrons moving in the presence of a random potential. Kubo[12] and Greenwood[13] have shown that the zero-temperature dc conductivity may be written as

$$\sigma_{\mu\nu} = \frac{\pi\hbar}{V} \left\langle \sum_{\alpha,\alpha'} \langle\alpha|j_\mu|\alpha'\rangle\langle\alpha'|j_\nu|\alpha\rangle \right\rangle \times \delta(E_F - E_\alpha)\delta(E_F - E_\alpha), \quad (2.13)$$

where j_μ is the current operator $(-ie\hbar/m)\partial/\partial r_\mu$, and V is the volume. The quantum states $|\alpha\rangle$ are the exact eigenfunctions of a particular configuration of the random potential and the large angle brackets indicate an average over configurations. By use of the Green function, defined as $G = [E - H]^{-1}$, which is related to the sum over states in Eq. (2.13) through

$$\sum_\alpha |\alpha\rangle\langle\alpha|\delta(E - E_\alpha) = -\frac{1}{\pi} \lim_{\eta\to 0} \mathrm{Im}G(E + i\eta). \quad (2.14)$$

Equation (2.13) can be written in the form

$$\sigma_{\mu\nu} = \frac{\pi\hbar}{V} \mathrm{Tr}\langle j_\mu \mathrm{Im}G(E_F) j'_\nu \mathrm{Im}G(E_F)\rangle. \quad (2.15)$$

Note that the Kubo formula requires an average over the product of two Green functions, $\langle GG\rangle$, rather than the product of the average of Green functions, $\langle G\rangle\langle G\rangle$. The error made when the former is approximated by the latter is, in general, quite serious. In the semiclassical limit this approximation is equivalent to the lifetime approximation or the neglect of vertex corrections. For the model of free electrons scattered by point scatterers which is considered in this paper, however, vertex corrections vanish because the scattering is isotropic.

D. Camblong-Levy theory

Camblong and Levy[14,15] derived an expression for the conductivity by making a semiclassical approximation to the Kubo formula. This expression will be derived in detail in Sec. III for a homogeneous system. For a general multilayer system the Camblong-Levy expression is a simple generalization of that result. The two-point con-

ductivity function which gives the current at plane z due to an electric field applied at point z' in the Camblong-Levy approximation is given by

$$\sigma_{zz}(z,z') = \frac{3}{2}\sigma_0\ell_0 \int_1^\infty \frac{dt}{t} e^{-\phi(z,z')t}, \tag{2.16}$$

and

$$\sigma_{xx}(z,z') = \frac{3}{4}\sigma_0\ell_0 \int_1^\infty dt \left(\frac{1}{t} - \frac{1}{t^3}\right) e^{-\phi(z,z')t}, \tag{2.17}$$

where ℓ_0 is the bulk mean free path, and $\phi(z,z')$ is given by an integral over the inverse of the local mean free path,

$$\phi(z,z') = \int_{z_<}^{z_>} dz'' \frac{1}{l(z'')}. \tag{2.18}$$

We shall see that the Camblong-Levy theory is closely related to the other semiclassical theories. Specifically, it is equivalent to Fuchs-Sondheimer with $p = 1$ for all interfaces. Since it is difficult to introduce a specularity coefficient $p \neq 1$ into the Camblong-Levy approach, they suggested that diffuse scattering at interfaces be treated by inserting an additional thin layer at the interface with a small mean free path.

E. Zhang-Levy-Fert theory

Zhang, Levy, and Fert[11] (ZLF) began with the Kubo formula for the FERPS model applied to multilayer systems in which the scattering rate is different in different layers. They made approximations which allowed them to obtain a simple expression for the local conductivity. The final expressions which ZLF give for the conductivity are relatively simple. The conductivity can be expressed in terms of a local lifetime,

$$\sigma_{xx}(z) = \frac{ne^2\tau(z)}{m}, \tag{2.19}$$

where the lifetime, $\tau(z)$, is obtained from an average over a z-dependent scattering rate, $\Delta(z)/\hbar$,

$$\frac{\hbar}{\tau(z)} = \frac{2}{\ell}\int dz' \Delta(z') E_1\left(\frac{|z-z'|}{\ell}\right). \tag{2.20}$$

An alternative version of the theory replaces the exponential integral in the above expression by $\exp(-|z - z'|/\ell)$. The overall mean free path ℓ appearing in Eq. (2.20) is defined in terms of an average scattering rate through

$$\ell = \frac{\hbar k_F}{m}\frac{\hbar L}{\int_0^L \Delta(z)dz}. \tag{2.21}$$

The Camblong-Levy theory can be described as a theory based on a local mean free path, while the Zhang-Levy-Fert theory can be described as a theory based on a local scattering rate. It is probably not surprising that the Zhang-Levy-Fert theory works best in the limit in which the layers are very thin, because in that limit the local scattering rate is the same as the average scattering rate.

III. EXACT LAYER CONDUCTIVITY FOR A HOMOGENEOUS FREE ELECTRON SYSTEM

The Hamiltonian in the FERPS model is $H = -\frac{\hbar^2}{2m}\nabla^2 + \Sigma$, where Σ is the complex self-energy which (in this section) we take to be independent of position. The constant real part of the self-energy simply gives an overall shift to the energy zero and can be neglected. The (negative) imaginary part of the self-energy leads to a broadening of the energy levels and to a finite lifetime for the eigenstates. The problem of a position dependent self-energy is more complex and will be considered in later sections.

The single particle Green function, which describes the propagation of the wave function amplitude, satisfies

$$\left[\frac{\hbar^2}{2m}\nabla^2 + E - \Sigma\right] G(\mathbf{r},\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}'), \tag{3.1}$$

and for a homogeneous system is given by

$$G(\mathbf{r},\mathbf{r}') = \frac{1}{(2\pi)^3}\int d^3k \frac{e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}}{E - \Sigma - \frac{\hbar^2k^2}{2m}} = \frac{2m}{\hbar^2}\frac{e^{i\kappa|\mathbf{r}-\mathbf{r}'|}}{4\pi|\mathbf{r}-\mathbf{r}'|}, \tag{3.2}$$

where $\kappa = \sqrt{2m(E-\Sigma)}/\hbar$.

According to the Kubo-Greenwood linear response formula, which we take to be exact for our model, the current for a single spin component at point $\mathbf{r}$ in direction μ is related to the ν component of the applied field at point $\mathbf{r}'$ through,

$$J_\mu(\mathbf{r}) = \int d^3r' \sigma_{\mu\nu}(\mathbf{r},\mathbf{r}') E_\nu(\mathbf{r}'), \tag{3.3}$$

where the nonlocal conductivity, $\sigma_{\mu\nu}(\mathbf{r},\mathbf{r}')$, is given from Eq. (2.15) by

$$\sigma_{\mu\nu}(\mathbf{r},\mathbf{r}') = \frac{\hbar}{\pi} j_\mu \mathrm{Im}G(\mathbf{r},\mathbf{r}') j'_\nu \mathrm{Im}G(\mathbf{r}',\mathbf{r}). \tag{3.4}$$

For a homogeneous, isotropic system, the expression for the conductivity can be simplified by integrating over $\mathbf{r}'$ and averaging over $\mathbf{r}$ and directions (μ). Thus, $\mathbf{J} = \sigma_0\boldsymbol{\mathcal{E}}$, where

$$\begin{aligned}\sigma_0 &= \frac{1}{3V}\sum_\mu \int d^3r \int d^3r' \sigma_{\mu\mu}(\mathbf{r},\mathbf{r}') \\ &= -\frac{1}{3}\frac{e^2\hbar^3}{\pi m^2 V}\int d^3r \int d^3r' \nabla \mathrm{Im}G(\mathbf{r},\mathbf{r}')\cdot\nabla'\mathrm{Im}G(\mathbf{r}',\mathbf{r}).\end{aligned} \tag{3.5}$$

We now use the Green function for the free electrons, Eq. (3.2), and setting $\mathbf{R} = \mathbf{r} - \mathbf{r}'$ and $|\mathbf{R}| = R$, we have

$$\sigma_0 = \frac{e^2\hbar^3}{3\pi m^2}\int d^3R [\nabla \mathrm{Im}G(R)]^2. \tag{3.6}$$

This integration can be performed exactly and yields,

$$\sigma_0 = \frac{e^2}{12\pi^2\hbar}\frac{\kappa_R^2}{\kappa_I}, \tag{3.7}$$

where κ_R and κ_I are the real and the imaginary parts of κ, respectively. In order to compare this result to the well-known formula, $\sigma = ne^2\tau/m$, where n is the carrier density and τ is the lifetime, we express the mean free path, ℓ as the Fermi velocity times the lifetime, $\ell = v_F\tau = \hbar k_F\tau/m$. We also write the carrier density, n, in terms of the volume of the Fermi sphere, $n = \frac{1}{8\pi^3}\frac{4}{3}\pi k_F^3$. This gives

$$\sigma_0 = \frac{e^2}{6\pi^2\hbar}k_F^2\ell. \tag{3.8}$$

Comparing this with Eq. (3.7), we can identify $\kappa_R = k_F$ and $\ell = 1/2\kappa_I$. The factor of $1/2$ comes from the fact that κ_I describes a decay rate for the electron *amplitude*, while $1/\ell$ is the decay rate for the electron *density*.

For layered systems which we assume to be homogeneous in two dimensions, we will need the layer dependent conductivity, $\sigma_{\mu\nu}(z,z')$ in order to determine the current distribution. It is obtained by integrating Eq. (3.4) over the $x'y'$ coordinates, and averaging over the xy coordinates. Thus

$$\sigma_{\mu\nu}(z,z') = -\frac{e^2\hbar^3}{\pi m^2 A}\int dxdy\int dx'dy'\nabla_\mu \mathrm{Im}G(\mathbf{r},\mathbf{r}')\nabla'_\nu \mathrm{Im}G(\mathbf{r}',\mathbf{r}), \tag{3.9}$$

where A is the area of the layers. Physically, $\sigma_{\mu\nu}(z,z')$ gives the contribution to the current at z due to an electric field applied at z'.

For a three dimensionally homogeneous system $\sigma_{\mu\nu}(z,z')$ depends only on $Z = |z-z'|$, and it is shown in Appendix A that

$$\begin{aligned}\sigma_{zz}(Z) = \sigma_0\frac{3\kappa_I}{2\kappa_R^2}\Big[&\kappa^2E_3(-2i\kappa Z) + \kappa^{*2}E_3(2i\kappa^* Z) + 2|\kappa|^2E_3(2\kappa_I Z)\\ &+\frac{2i\kappa}{Z}E_4(-2i\kappa Z) - \frac{2i\kappa^*}{Z}E_4(2i\kappa^* Z) + \frac{4\kappa_I}{Z}E_4(2\kappa_I Z)\\ &-\frac{1}{Z^2}E_5(-2i\kappa Z) - \frac{1}{Z^2}E_5(2i\kappa^* Z) + \frac{2}{Z^2}E_5(2\kappa_I Z)\Big],\end{aligned} \tag{3.10}$$

and

$$\begin{aligned}\sigma_{xx}(Z) = \sigma_0\frac{3\kappa_I}{4\kappa_R^2}\Big[&\kappa^2E_1(-2i\kappa Z) + \kappa^{*2}E_1(2i\kappa^* Z) + 2|\kappa|^2E_1(2\kappa_I Z)\\ &+\frac{2i\kappa}{Z}E_2(-2i\kappa Z) - \frac{2i\kappa^*}{Z}E_2(2i\kappa^* Z) + \frac{4\kappa_I}{Z}E_2(2\kappa_I Z)\\ &-\frac{1}{Z^2}E_3(-2i\kappa Z) - \frac{1}{Z^2}E_3(2i\kappa^* Z) + \frac{2}{Z^2}E_3(2\kappa_I Z)\Big] - \frac{1}{2}\sigma_{zz}(Z),\end{aligned} \tag{3.11}$$

where $E_n(x)$ is the exponential integral of nth order defined as

$$E_n(x) = \int_1^\infty \frac{e^{-xt}}{t^n}dt, \tag{3.12}$$

and σ_0 is the total bulk conductivity as given by Eq. (3.7).

The nonlocal layer dependent conductivities are shown in Fig. 1. There are obvious quantum oscillations for current and field perpendicular to the layers (σ_{zz}). The quantum oscillations are much smaller when field and current are parallel to the layers. For current and field perpendicular to the layers we have the surprising result that a field applied only within a vanishingly thin layer does not induce a current in that layer.

In a real system it is difficult to think of a probe smaller than an atom and it is unlikely that the applied field can change appreciably over such a distance, so in applications of the nonlocal conductivity formula it will generally be averaged over a distance, d, equal at least to the thickness of an atomic layer. Thus the layer conductivity, σ^{IJ}, which is the response in layer I due to a field in layer J, is obtained from the equation,

$$\sigma_{\mu\nu}^{IJ} = \frac{1}{d}\int_0^d dz\int_0^d dz'\sigma_{\mu\nu}(Z_{IJ}+z-z'), \tag{3.13}$$

where $Z_{IJ} = |I-J|d$. One of the integrals can be carried out analytically. Using

$$\int dzE_n(\alpha z) = -\frac{1}{\alpha}E_{n+1}(\alpha z), \tag{3.14}$$

and

$$\int dz\left[\frac{\alpha}{z}E_n(\alpha z) + \frac{1}{z^2}E_{n+1}(\alpha z)\right] = -\frac{1}{z}E_{n+1}(\alpha z), \tag{3.15}$$

we can evaluate the integral

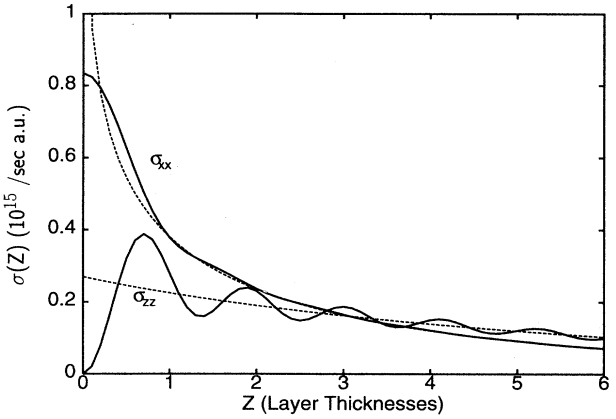

FIG. 1. Quantum and semiclassical nonlocal layer conductivity as a function of layer separation, Z, for a homogeneous free-electron system. Solid lines are the quantum conductivity. Dashed lines are the semiclassical approximation. The lattice constant is that of copper (0.3615 nm). The Fermi momentum corresponds to 0.5 electrons per spin channel. Z is measured in terms of the thickness of (111) layers of copper (0.209 nm). $\sigma(Z)$ is measured in units of 10^{15}/sec a.u. where 1 a.u.=0.0529 nm. The mean free path is ten layer thicknesses.

$$\sigma_{\mu\nu}^{IJ}(z) \equiv \int_0^d dz' \sigma_{\mu\nu}(Z_{IJ}+z-z') = \int_{z+Z_{IJ}-d}^{z+Z_{IJ}} dZ \sigma_{\mu\nu}(Z). \tag{3.16}$$

The result is simple but too long to include here. The final step in the calculation of σ_{zz}^{IJ} and σ_{xx}^{IJ} is the integration of $\sigma_{zz}^{IJ}(z)$ and $\sigma_{xx}^{IJ}(z)$ from Z_{IJ} to $Z_{IJ}+d$, which can be done numerically. Calculated results for σ_{zz}^{IJ} and σ_{xx}^{IJ} are shown in Fig. 2 together with their semiclassical limits which are obtained in the next section.

IV. THE SEMICLASSICAL LIMIT OF THE KUBO FORMULA FOR FREE ELECTRONS

The semiclassical limit of Eqs. (3.10) and (3.11) is obtained by replacing these expressions with expressions without quantum oscillations and which represent an average over the quantum expressions. There are nine terms in the expression Eq. (3.10) for $\sigma_{zz}(Z)$. The six terms containing exponential integrals with complex arguments, ($-2i\kappa Z$ or $2i\kappa^* Z$), are oscillatory functions of Z and should be replaced using the following rules. First, we replace an oscillatory exponential function with a function that has the same decay length and overall volume integral, but without quantum oscillations,

$$e^{i2\kappa Z t} \to \frac{i\kappa_I}{\kappa} e^{-2\kappa_I Z t}. \tag{4.1}$$

To obtain the semiclassical limit of the terms involving $1/Z$, we integrate the above equation over t and find

$$\frac{1}{Z} e^{i2\kappa Z t} \to \frac{1}{Z} e^{-2\kappa_I Z t}. \tag{4.2}$$

Finally, we integrate over t once more to obtain

$$\frac{1}{Z^2} e^{i2\kappa Z t} \to \frac{-i\kappa}{\kappa_I Z^2} e^{-2\kappa_I Z t}. \tag{4.3}$$

Using the above substitutions in Eq. (3.10), we find that the terms with E_4 and E_5 cancel exactly, and the terms with E_3 yield

$$\sigma_{zz}^{\rm sc}(Z) = 3\sigma_0 \kappa_I \, E_3(2\kappa_I Z). \tag{4.4}$$

Similarly, for the semiclassical limit of Eq. (3.11), we obtain

$$\sigma_{xx}^{\rm sc}(Z) = \frac{3}{2}\sigma_0 \kappa_I [E_1(2\kappa_I Z) - E_3(2\kappa_I Z)]. \tag{4.5}$$

Figure 1 shows the functions $\sigma_{zz}(Z)$, $\sigma_{xx}(Z)$, $\sigma_{zz}^{\rm sc}(Z)$, and $\sigma_{xx}^{\rm sc}(Z)$ for parameters corresponding to a free-electron system with one electron per atom and the lattice constant of copper. The major differences between the quantum and semiclassical expressions are the oscillations in the quantum case, which are missing in the semiclassical approximation. These oscillations are much more apparent for $\sigma_{zz}(Z)$ than for $\sigma_{xx}(Z)$. The quantum and semiclassical functions also behave differently in the limit $Z \to 0$. The quantum function, $\sigma_{zz}(Z)$ vanishes quadratically as $Z \to 0$, while the semiclassical version goes linearly to $3\kappa_I\sigma_0/4$. On the other hand, the quantum function $\sigma_{xx}(Z)$ assumes a finite value at the origin, while the semiclassical version diverges as $-1.5\sigma_0\kappa_I[\ln(2\kappa_I Z)+\gamma]$ as $Z \to 0$.

The quantum and semiclassical versions of the free-electron two point conductivities are much more similar when they are averaged over a length scale corresponding to the thickness of an atomic layer. Integration over z and z' using Eq. (3.13) gives

$$\sigma_{zz}^{IJ\rm sc} = \frac{3\sigma_0}{4\kappa_I d}\{E_5[2\kappa_I(Z_{IJ}-d)] - 2E_5(2\kappa_I Z_{IJ}) + E_5[2\kappa_I(Z_{IJ}+d)]\}, \tag{4.6}$$

and

$$\sigma_{xx}^{IJ\rm sc} = \frac{3\sigma_0}{8\kappa_I d}\{E_3[2\kappa_I(Z_{IJ}-d)] - 2E_3(2\kappa_I Z_{IJ}) + E_3[2\kappa_I(Z_{IJ}+d)]\} - \frac{1}{2}\sigma_{zz}^{IJ\rm sc}, \tag{4.7}$$

for $I \neq J$, and

$$\sigma_{zz}^{II\rm sc} = \sigma_0\left[1 - \frac{3}{8\kappa_I d} + \frac{3}{2\kappa_I d}E_5(2\kappa_I d)\right] \tag{4.8}$$

and

$$\sigma_{xx}^{II\rm sc} = \sigma_0\left\{1 - \frac{3}{16\kappa_I d} + \frac{3}{4\kappa_I d}[E_3(2\kappa_I d) - E_5(2\kappa_I d)]\right\} \tag{4.9}$$

for the on-site terms. Figure 2 compares the integrated

versions of the conductivity functions. Although most of the differences between the quantum and semiclassical results have been washed out by the averaging, discernible differences remain, especially for small values of Z.

A. Camblong-Levy approach

The results obtained above can be compared with the results of previous semiclassical theories. Camblong and Levy[14,15] obtained Eqs. (4.4) and (4.5) using an approach in which the semiclassical average over the product of Green functions in the Kubo formula is performed before the integration over space. We outline their approach below.

The Green function can be represented either in real space or in reciprocal space. For layered systems which are homogeneous in two dimensions it is sometimes convenient to represent the Green function using a hybrid representation; reciprocal space for the variation parallel to the layers and real space for the variation perpendicular. Thus,

$$G(\mathbf{r},\mathbf{r}') = \frac{1}{(2\pi)^2}\int dk_x dk_y e^{i[k_x(x-x')+k_y(y-y')]}\frac{1}{2\pi}\int dk_z \frac{e^{ik_z(z-z')}}{E-\Sigma-\frac{\hbar^2}{2m}(k_x^2+k_y^2+k_z^2)}$$
$$= \frac{1}{(2\pi)^2}\int d^2k_\parallel e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel}G(k_\parallel,z,z'), \tag{4.10}$$

where $G(k_\parallel,z,z') = \frac{2m}{\hbar^2}e^{i\bar{k}Z}/2i\bar{k}$ with $\bar{k} = \sqrt{2m(E-\Sigma)/\hbar^2 - k_\parallel^2}$. The Kubo formula for $\sigma_{zz}(Z)$ in terms of $G(k_\parallel,z,z')$ is

$$\sigma_{zz}(Z) = \frac{e^2\hbar^3}{\pi m^2}\int d^2\mathbf{R}_\parallel \frac{1}{(2\pi)^2}\int d^2k_\parallel \frac{1}{(2\pi)^2}\int d^2k'_\parallel e^{i(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)\cdot\mathbf{R}_\parallel}\frac{\partial}{\partial Z}\mathrm{Im}G(k_\parallel,Z)\frac{\partial}{\partial Z}\mathrm{Im}G(k'_\parallel,Z). \tag{4.11}$$

To calculate $\sigma_{zz}(Z)$, we need $\frac{\partial}{\partial Z}\mathrm{Im}G(k_\parallel,Z)$, which is given by

$$\frac{\partial}{\partial Z}\mathrm{Im}G(k_\parallel,Z) = \frac{m}{\hbar^2}\sin\bar{k}_R Z e^{-\bar{k}_I Z}, \tag{4.12}$$

where $\bar{k}_R$ and $\bar{k}_I$ are the real and the imaginary parts of $\sqrt{2m(E-\Sigma)/\hbar^2 - k_\parallel^2}$, respectively. Averaging the square of this function and inserting into the Kubo formula we have

$$\sigma_{zz}(Z) = \frac{e^2}{4\pi^2\hbar}\int k_\parallel dk_\parallel e^{-2\bar{k}_I Z}. \tag{4.13}$$

If we assume that the mean free path is much longer than the electron wavelength, i.e., $\kappa_I \ll \kappa_R$, we can use $\bar{k}^2 = \frac{2m}{\hbar^2}(E-\Sigma) - k_\parallel^2 \approx \kappa_R^2 + 2i\kappa_R\kappa_I - k_\parallel^2 = \bar{k}_R^2 + 2i\bar{k}_R\bar{k}_I$ to obtain

$$\sigma_{zz}(Z) = \frac{e^2}{4\pi^2\hbar}\int_0^{\kappa_R} k_\parallel dk_\parallel e^{-2Z\kappa_R\kappa_I/\bar{k}_R}. \tag{4.14}$$

Setting $t = \kappa_R/\bar{k}_R$ and changing variables, $k_\parallel dk_\parallel = \kappa_R^2 dt/t^3$ we obtain

$$\sigma_{zz}(Z) = 3\sigma_0\kappa_I\int_1^\infty \frac{dt}{t^3}e^{-2Z\kappa_I t}. \tag{4.15}$$

Similarly, for the parallel semiclassical conductivity, we have

$$\sigma_{xx}(Z) = \frac{e^2}{4\pi^2\hbar}\int_0^{\kappa_R} k_\parallel dk_\parallel \frac{k_\parallel^2}{\bar{k}^2}e^{-2Z\kappa_R\kappa_I/\bar{k}_R} \tag{4.16}$$

from which we obtain

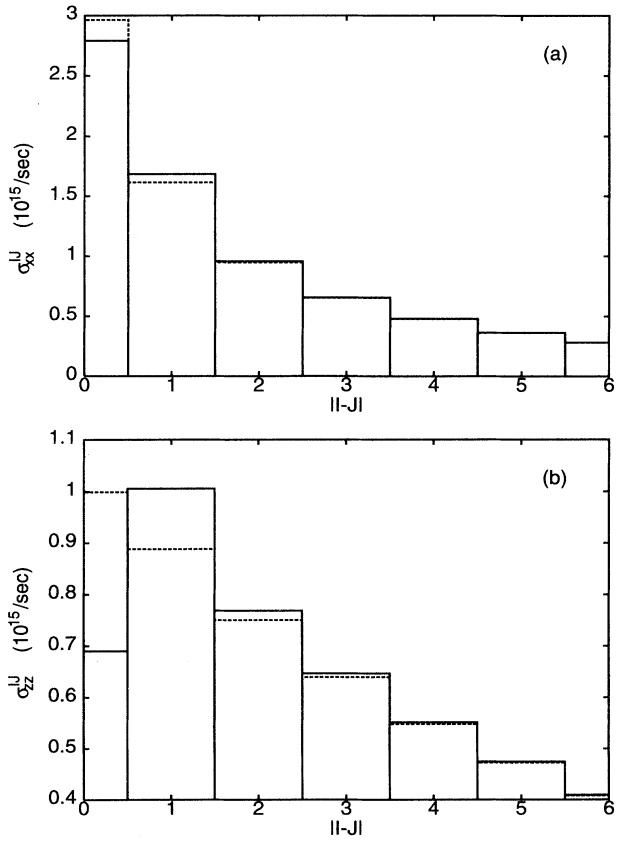

FIG. 2. Interlayer conductivity for a homogenous free-electron system (a) parallel to the layers and (b) perpendicular to the layers. The parameters are the same as for Fig. 1. Solid lines are the quantum conductivity, dashed lines are the semiclassical approximation.

$$\sigma_{xx}(Z) = \frac{3}{2}\sigma_0\kappa_I \int_1^\infty dt \left(\frac{1}{t} - \frac{1}{t^3}\right) e^{-2Z\kappa_I t}. \quad (4.17)$$

These two-point conductivities are the same as those given by Eqs. (4.4) and (4.5); however, the derivation of Eqs. (4.4) and (4.5) was somewhat cleaner since it was not necessary to assume that $\kappa_I \ll \kappa_R$ in taking the semiclassical limit of the exact result.

B. Chambers-Pippard approach

Next, we consider the Chambers-Pippard approach. Using Eqs. (2.11) and (2.12) we can write the current as

$$\mathbf{J} = \frac{e}{(2\pi)^3} \int d^3k \mathbf{v_k} \frac{\partial f_0}{\partial E} \int_0^\infty dR \frac{e\boldsymbol{\mathcal{E}}(\mathbf{R})\cdot\mathbf{R}}{R} e^{-R/\ell}, \quad (4.18)$$

where $\mathbf{R} = \mathbf{r}' - \mathbf{r}$. The derivative of the Fermi function reduces the integral over $\mathbf{k}$ to an integral over the Fermi surface and since $\mathbf{R}$ and $\mathbf{v_k}$ are in the same direction, we have

$$\mathbf{J} = \frac{e^2}{8\pi^3\hbar} \oint dS_k \int_0^\infty dR \frac{\mathbf{R}\boldsymbol{\mathcal{E}}(\mathbf{R})\cdot\mathbf{R}}{R^2} e^{-R/\ell}. \quad (4.19)$$

For free electrons, the element of Fermi surface area dS_k is simply $k_F^2 d\Omega$, so that

$$\mathbf{J} = \frac{e^2k_F^2}{8\pi^3\hbar} \int d^3R \frac{\mathbf{R}\,\boldsymbol{\mathcal{E}}(\mathbf{R})\cdot\mathbf{R}}{R^4} e^{-R/\ell}. \quad (4.20)$$

If we view the integrand of the above equation as the current response at the point $\mathbf{r}$ due to the field at the point $\mathbf{r}'$, the layer conductivity is then given by

$$\sigma_{zz}(Z) = 2\pi \int R_\parallel dR_\parallel \frac{J_z(\mathbf{R})}{\mathcal{E}_z} = \frac{e^2k_F^2}{4\pi^2\hbar} \int R_\parallel dR_\parallel \frac{Z^2}{R^4} e^{-R/\ell}, \quad (4.21)$$

and

$$\sigma_{xx}(Z) = 2\pi \int R_\parallel dR_\parallel \frac{J_x(\mathbf{R})}{\mathcal{E}_x} = \frac{e^2k_F^2}{4\pi^2\hbar} \int R_\parallel dR_\parallel \frac{1}{2}\frac{{R_\parallel}^2}{R^4} e^{-R/\ell}. \quad (4.22)$$

For a system in which the Fermi surface is not spherical, the factor k_F^2 is replaced by the inverse of the Gaussian curvature of the Fermi surface, which will vary as a function of angle. Thus, this approach (like the Boltzmann equation) can be applied to real solids because the band structure can be properly taken into account through the shape of the Fermi surface. Later we shall see that this theory has the additional advantage that it can be correctly generalized to a system, where the rate of scattering is inhomogeneous, although the theory as it was originally developed was for a homogeneous scattering rate but an inhomogeneous applied field.

Using $R^2 = {R_\parallel}^2 + Z^2$, we have

$$\sigma_{zz}(Z) = 3\sigma_0\kappa_I \int_1^\infty \frac{dt}{t^3} e^{-tZ/\ell}, \quad (4.23)$$

and

$$\sigma_{xx}(Z) = \frac{3\sigma_0\kappa_I}{2} \int_1^\infty dt \left(\frac{1}{t} - \frac{1}{t^3}\right) e^{-tZ/\ell}, \quad (4.24)$$

where we made a change of variable, $t = \sqrt{1 + {R_\parallel}^2/Z^2}$. Again, these are exactly the semiclassical limits of the Kubo formula for $\sigma_{xx}^{sc}(Z)$ and $\sigma_{zz}^{sc}(Z)$, Eqs. (4.4) and (4.5).

C. Fuchs-Sondheimer approach

In this subsection, we consider the Fuchs-Sondheimer approach[7,8] introduced in Sec. II A. In order to compare with the exact results given in the previous sections, we need to generalize the Fuchs-Sondheimer approach to give two point conductivities for multilayers. To do this we write the external field as $E = \sum_J E_J$, where E_J is a uniform field within a layer J, but zero elsewhere. This enables us to decompose the solution for layer I, $g^I(\mathbf{v}, z)$, where z lies within the layer I, into a sum over the contributions due to E_J, from layers J,

$$g^I(\mathbf{v}, z) = \sum_J g^{IJ}(\mathbf{v}, z). \quad (4.25)$$

The contribution to the current, $\mathbf{J}^{IJ}$, is given by

$$\mathbf{J}^{IJ} = \frac{e}{(2\pi)^3} \int d^3k \mathbf{v_k} [g_+^{IJ}(\mathbf{v_k})\Theta(v_z(\mathbf{k})) + g_-^{IJ}(\mathbf{v_k})\Theta(-v_z(\mathbf{k}))]. \quad (4.26)$$

In Appendix B, we show that for the current perpendicular to the plane (CPP) the conductivity is given by

$$\sigma_{zz}^{II} = \sigma_0 \left[1 - \frac{3}{4}\frac{\ell}{d} + 3\frac{\ell}{d} E_5\left(\frac{d}{\ell}\right)\right], \quad (4.27)$$

and for the out-of-plane terms,

$$\sigma_{zz}^{IJ} = \sigma_0 \frac{3}{2}\frac{\ell}{d}\left[E_5\left(\frac{(|I-J|-1)d}{\ell}\right) - 2E_5\left(\frac{|I-J|d}{\ell}\right) + E_5\left(\frac{(|I-J|+1)d}{\ell}\right)\right], \quad (4.28)$$

where σ_0 is the total conductivity of the system as given by Eq. (3.8).

The current in-plane (CIP) case can be solved in a similar manner. We obtain

$$\sigma_{xx}^{II} = \sigma_0 \left\{1 - \frac{3}{8}\frac{\ell}{d} + \frac{3}{2}\frac{\ell}{d}\left[E_3\left(\frac{d}{\ell}\right) - E_5\left(\frac{d}{\ell}\right)\right]\right\}, \quad (4.29)$$

and for the out-of-plane terms,

$$\sigma_{xx}^{IJ} = \sigma_0 \frac{3}{4}\frac{\ell}{d}\left[E_3\left(\frac{(|I-J|-1)d}{\ell}\right) - 2E_3\left(\frac{|I-J|d}{\ell}\right) + E_3\left(\frac{(|I-J|+1)d}{\ell}\right) - E_5\left(\frac{(|I-J|-1)d}{\ell}\right) + 2E_5\left(\frac{|I-J|d}{\ell}\right) - E_5\left(\frac{(|I-J|+1)d}{\ell}\right)\right]. \quad (4.30)$$

Remembering that the mean free path is given by $\ell = 1/2\kappa_I$, we observe that these equations agree with the semiclassical limit of the results obtained from the Kubo formula, Eqs. (4.6), (4.7), (4.8), and (4.9).

From the results presented in this and the preceding section, we conclude that all three semiclassical approaches give identical results in the FERPS model for the two-point conductivity tensor and that these results agree with the semiclassical limit of the quantum result. We observe further from Figs. 1 and 2 that the differences between the quantum and semiclassical expressions for the two-point conductivity tensor are relatively small for metallic systems when averaged over a layer thickness. One exception to this conclusion would be for a system with a low value for k_F, e.g., a semimetal. For such a system the small $k_F d$ regime in which quantum oscillations are important should be accessible.

V. THIN FILMS

In this section, we use the free-electron model to compare various approaches to the calculation of the conductivity of thin metallic films. This study has relevance to recent work on the GMR, because techniques originally devised for treating the effects of surfaces on the conductivity of films have been pressed into service to treat the effects of interfacial scattering in multilayers. The effect of surface scattering on the transport properties of thin metallic films has been studied extensively. For many years, the semiclassical theory developed by Fuchs[7] and Sondheimer[8] was thought to be adequate for describing the effect of the surface. More recently it has been recognized that the specularity parameter p, which was assumed to be a constant in the original Fuchs-Sondheimer theory, is necessarily dependent on the angle of incidence of the electron on the surface. Modifications of the theory have been proposed to include this effect.[19,20]

Fuchs-Sondheimer theory[7,8,21] applied to a thin film with $p \neq 1$ predicts a logarithmic divergence of the conductivity as d/ℓ, the ratio of thickness to mean free path, goes to zero. Although early experiments[22] seemed to agree with this prediction, recent experiments on Pt (Ref. 23) and $CoSi_2$ (Ref. 24) films show considerable disagreement with this result. Recently, it has been shown[25,26] that this disagreement is a quantum effect. Here we confirm this result, and show that the dominant contribution arises from the zero-point motion of the electrons in the perpendicular direction. We also show that a semiclassical limit of the quantum result can be obtained, which differs from the Fuchs-Sondheimer result in that it properly accounts for the electronic zero-point motion and has an angle dependent specularity parameter. The results of this theory appear to agree well with experiments. On the other hand, the Fuchs-Sondheimer theory with a constant p does not agree with experiment even with the zero-point motion included. The use of an angle dependent p allows us to unify the Fuchs-Sondheimer theory with that of Chambers-Pippard and with the quantum results from the Kubo formula.

A. Quantum solution for thin films

We consider a thin film that extends infinitely with uniform electronic potential in the x and y directions and has a thickness d in the z direction. We assume that the electronic self-energy $\Sigma(z)$ depends only on z. The partial Green function, $G(k_\parallel; z, z')$, defined in Sec. IV A satisfies

$$\left[E + \frac{\hbar^2}{2m}\left(\frac{\partial^2}{\partial z^2} - k_\parallel^2\right) - \Sigma(z)\right] G(k_\parallel; z, z') = \delta(z - z'). \quad (5.1)$$

Let us assume initially that the imaginary part of the self-energy is independent of z, i.e., $\Sigma(z) = v(z) + \Sigma$, where $v(z)$ is real and Σ is imaginary. The Green function for this thin film can then be written as

$$G(k_\parallel; z, z') = \sum_n \frac{\phi_n(z)\phi_n^*(z')}{E - \Sigma - \epsilon_n - \frac{\hbar^2 k_\parallel^2}{2m}}, \quad (5.2)$$

where the functions $\phi_n(z)$ are solutions to the one-dimensional Schrödinger equation,

$$\left[-\frac{\hbar^2}{2m}\frac{d^2}{dz^2} + v(z)\right]\phi_n(z) = \epsilon_n \phi_n(z). \quad (5.3)$$

The conductivity parallel to the film is then given by the Kubo formula,

$$\sigma_{xx} = -\frac{1}{8\pi^3}\frac{e^2\hbar^3}{m^2 d}\int_0^d dz \int_0^d dz' \int d^2 k_\parallel \times k_\parallel^2 \mathrm{Im}G(k_\parallel; z, z')\mathrm{Im}G(k_\parallel; z', z). \quad (5.4)$$

It is shown in Appendix C that the total conductivity is equal to

$$\sigma_{xx} = \frac{e^2}{4\pi^2\hbar d}\left\{\sum_{n=1}^{N}\frac{\alpha_n}{\eta}\pi + \sum_{n=1}^{\infty}\left[1 - \frac{\alpha_n}{\eta}\arctan\frac{\eta}{\alpha_n}\right]\right\}, \quad (5.5)$$

where $\alpha_n = 2m(E - \epsilon_n)/\hbar^2$, $\eta = 2mi\Sigma/\hbar^2$, and N is the largest integer for which $\epsilon_N \leq E$. The second term represents a contribution from the smearing of the energy bands due to disorder. Trivedi and Ashcroft[26] obtained a similar result for the special case of an infinite square well and constant $v(z)$. They did not, however, include

the broadening of the energy levels due to disorder and so their result does not include the second term of Eq. (5.5). This term is small if the scattering is weak so that their result is valid for weak scattering. Equation (5.5) is valid, however, for arbitrarily strong scattering if it is uniform throughout the film.

In Fig. 3, we plot the conductivity as a function of the film thickness for an infinite square-well and uniform scattering rate. The conductivity is calculated from Eq. (5.5), where the eigenenergies are taken to be those of an infinite square well potential, and the imaginary part of the energy is a constant. The quantum size effect, which cuts off the conductivity abruptly for $d < \pi/k_F$, and the quantum oscillations, which are manifest as cusps at $d = n\pi/k_F$, are clearly evident. It should be remembered, of course, that it is difficult to observe these quantum effects since in most metals π/k_F is approximately the thickness of a single atomic layer. Semimetals with a very small effective k_F might be good candidates for showing these effects.

In order to describe real metal films which have thicknesses small compared to the electron mean free path, it is necessary to confront the problem of electron scattering from the surfaces. Surface scattering is usually treated following Fuchs and Sondheimer by using a parameter p to describe the fraction of electrons that are specularly reflected from a surface. The validity of this approach and its justification in terms of a quantum theory have been difficult to assess. We prefer to address scattering off of surfaces, boundaries, and interfaces similarly to bulk scattering, i.e., we model such scattering by assuming the existence of a thin region with an extremely high scattering rate. We shall see that this approach leads naturally to the concept of a mean free path that depends on the direction of the electron velocity. We shall also show the relation between this angle dependent mean free path and the parameter p.

If the thickness of the thin surface regions is small compared to the mean free path in these regions so that the wave functions are not affected significantly by these regions, perturbation theory can be used to obtain the inverse lifetimes needed in Eq. (5.5). In Appendix D, we use perturbation theory to show that the lifetime of electronic eigenstates in a thin film with very thin regions of thickness, Δd, of stronger scattering at the boundaries has the form

$$\frac{1}{\tau_n} = \frac{1}{\tau_0}(1 + \xi k_z^2), \tag{5.6}$$

where $k_z = n\pi/d$, and ξ is the coefficient that describes the roughness of the surfaces and is a function of k_F, d, and Δd. The zeroth order term in ξ is proportional to $1/d$. The contributions to ξ in higher orders of $1/k_F d$ are significant for film thickness $d \approx \pi/k_F$. The constant τ_0 is the maximum lifetime, defined by Eq. (D5). By examining Eq. (D4) one can see that Eq. (5.6) with an adjustable coefficient, ξ, will generally be a reasonable representation for the variation of the lifetime even when the nonuniform scattering is not confined to the boundary region.

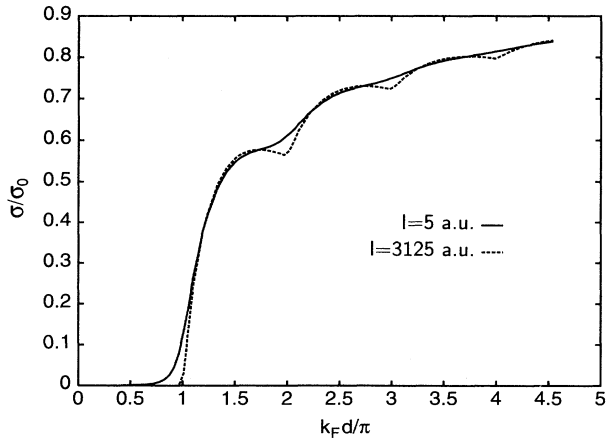

FIG. 3. Conductivity for a thin film with a square well potential and uniform random scattering. (1 a.u. = 0.529 Å.)

B. The continuum limit

The continuum limit of Eq. (5.5) is taken by assuming the film to be sufficiently thick that $k_z = n\pi/d$ can be treated as a continuous variable. We emphasize two important points about the continuum limit. First, Eq. (5.6) requires that η in Eq. (5.5) depend on the momentum in the z direction in order to account for surface scattering. Second, the integral over k_z which replaces the sum over n in the continuum limit should have a lower limit of $k_0 = \pi/d$, i.e., the smallest allowable wave vector in the z direction. It cannot be replaced by zero in the limit of long mean free path because in that limit the contribution from small k_z becomes singular. Now, if we further assume that on average the electronic potential is zero, i.e., $\langle v(z)\rangle = 0$ within the thin film, we can approximate the energy levels by $\epsilon_n = \hbar^2\langle k_z\rangle^2/2m$. Since the second term in Eq. (5.5) becomes negligible in the continuum limit, we have

$$\sigma_{xx} = \frac{e^2}{4\pi^2\hbar}\int_{k_0}^{k_F} dk_z \frac{k_F^2 - k_z^2}{\eta(k_z)}. \tag{5.7}$$

Using $\eta = k_F/\ell$ and making a change of variable, $t = k_F/k_z$, we obtain

$$\sigma_{xx} = \frac{e^2 k_F^2}{4\pi^2\hbar}\int_1^{k_F/k_0} dt \left(\frac{1}{t^2} - \frac{1}{t^4}\right)\ell(t). \tag{5.8}$$

The physical content of Eq. (5.8) is that the continuum limit of the quantum expression for the conductivity of a film is the same as the semiclassical limit of the expression for the bulk conductivity with two modifications: (1) The mean free path must depend on the direction of the electron relative to the normal to the surface, $[\theta = \cos^{-1}(1/t)]$. (2) There is a minimum value to the electron momentum perpendicular to the film surface, $k_0 = \pi/d$.

We can understand modification (1) from arguments based on semiclassical theory. Since we model the sur-

face scattering by a thin region with high scattering rate, the conductivity of our film is the same as that for an infinite periodic multilayer with regions of high scattering rate spaced at an interval equal to the film thickness, d. Then, since the electron moves on the Fermi surface of this periodic system rather than on a spherical Fermi surface, its mean free path will vary with k_z. This variation can be accommodated within Chambers-Pippard theory by making the mean free path angle dependent in Eq. (4.24),

$$\sigma_{xx}(Z) = \frac{3\sigma_0\kappa_I}{2}\int_1^\infty dt\left(\frac{1}{t}-\frac{1}{t^3}\right)e^{-tZ/\ell(t)}. \tag{5.9}$$

Integrating Eq. (5.9) over Z gives us

$$\sigma_{xx} = \frac{e^2k_F^2}{4\pi^2\hbar}\int_1^\infty dt\left(\frac{1}{t}-\frac{1}{t^3}\right)\frac{\ell(t)}{t}, \tag{5.10}$$

which is identical to the continuum limit of the quantum theory, Eq. (5.8), except for the difference in the upper limit of the integral over t.

Modification (2), the nonvanishing k_0, is an electronic "zero-point motion" in the z direction which arises from the finite thickness of the film. We will show by comparison with experiment in Sec. V D that this is the dominant quantum effect for thin films. It can be easily incorporated into a semiclassical theory by changing the upper limit of t from ∞ to k_F/k_0, or equivalently, changing the lower limit of $\cos\theta$ of the electron velocity from 0 to k_0/k_F.

The specific form of the dependence of $\ell(t)$ on t can be obtained by considering Eq. (5.6), which implies a quadratic dependence,

$$\frac{2m}{\hbar^2}i\Sigma = \eta_0 + \xi_2\langle k_z^2\rangle. \tag{5.11}$$

Equivalently, the mean free path has the form,

$$\ell(t) = \frac{\ell_0}{1+\xi_2\ell_0k_F/t^2}, \tag{5.12}$$

where we have used $i\Sigma = \hbar^2k_F/2m\ell$, $\eta_0 = k_F/\ell_0$, and $t = k_F/k_z$. This allows us to integrate Eq. (5.8) and leads to

$$\sigma_{xx} = \frac{e^2k_F}{4\pi^2\hbar\xi_2}[f(k_F)-f(k_0)], \tag{5.13}$$

where

$$f(k) = \sqrt{\ell_0k_F\xi_2}\left(1+\frac{1}{\ell_0k_F\xi_2}\right)\arctan\left(\frac{k}{k_F}\sqrt{\ell_0k_F\xi_2}\right) - \frac{k}{k_F}. \tag{5.14}$$

Note that if in Eq. (5.13), we take $k_0 = 0$ and then take the limit of $\ell_0 \to \infty$ the conductivity diverges as $\sqrt{\ell_0}$, instead of the well-known logarithmic divergence[7,8,21] that arises in Fuchs-Sondheimer theory for $p \neq 1$. This square root divergence arises from the quadratic k_z dependence of the mean free path in Eq. (5.11). In order to obtain the logarithmic divergence it is necessary to assume a linear k_z dependence,

$$\frac{2m}{\hbar^2}i\Sigma = \eta_0 + \xi_1\langle k_z\rangle, \tag{5.15}$$

or equivalently, for the mean free path,

$$\ell(t) = \frac{\ell_0}{1+\xi_1\ell_0/t}. \tag{5.16}$$

Note that Eq. (5.15) represents an unphysical assumption because it requires a self-energy that is nonanalytic at $k_z = 0$. However, we will show later that it is the only way to produce a semiclassical limit that agrees with the Fuchs-Sondheimer theory with a constant $p < 1$. In fact, we will also show that the parameters ξ_1 and p are connected by a simple relationship.

Using Eq. (5.16) in Eq. (5.8) we have

$$\sigma_{xx} = \frac{e^2k_f^2}{4\pi^2\hbar\xi_1}[g(k_F)-g(k_0)], \tag{5.17}$$

where

$$g(k) = \ln\left(1+\xi_1\ell_0\frac{k}{k_F}\right)\left(1-\frac{1}{\xi_1^2\ell_0^2}\right) + \frac{k}{k_F}\left(-\frac{1}{2}+\frac{1}{\xi_1\ell_0}\right). \tag{5.18}$$

In order to compare to the Fuchs-Sondheimer theory, we set $k_0 = 0$ and take the limit of $\ell_0 \to \infty$, and obtain

$$\sigma_{xx} = \frac{e^2k_F^2}{4\pi^2\hbar\xi_1}\ln\xi_1\ell_0. \tag{5.19}$$

It is interesting to compare the above results with the limit of Eq. (5.13), which when $\ell_0 \to \infty$, gives

$$\sigma_{xx} = \frac{e^2k_F}{4\pi^2\hbar\xi_2}\left(\frac{k_F}{k_0}+\frac{k_0}{k_F}-2\right). \tag{5.20}$$

If $k_F \gg k_0$, we have

$$\sigma_{xx} = \frac{e^2k_F^2}{4\pi^2\hbar\xi_2k_0} = \frac{e^2k_F^2}{6\pi^2\hbar}\frac{3d}{2\pi\xi_2}, \tag{5.21}$$

which shows that there is no divergence if k_0 is not zero. For the special case of $\xi_1 = 0$, $\xi_2 = 0$, and $k_F \gg k_0$. Both Eqs. (5.13) and (5.17) give the correct bulk conductivity, $\sigma_{xx} = e^2k_F^2\ell_0/6\pi^2\hbar$.

C. Generalized Fuchs-Sondheimer theory of surface roughness

If we generalize the Fuchs-Sondheimer theory for a thin film to allow for an angle dependent specularity parameter, $p(t)$, the distribution function will have the following boundary conditions:

$$g_+(t,z=0) = p(t)g_-(t,z=0),$$
$$g_-(t,z=d) = p(t)g_+(t,z=d), \tag{5.22}$$

where $t = 1/|\cos\theta|$, θ is the angle of incidence at the boundaries, and $g_\pm$ are the deviation of the distribution functions for electrons traveling in + (−) directions. The solution to the semiclassical Boltzmann equation with the above conditions is

$$g_\pm(t,z) = \frac{e\tau E}{m}\frac{\partial f_0}{\partial v_\parallel}\left[1 - \frac{1-p(t)}{1-p(t)e^{-td/\ell}}e^{-t|z-z_\pm|/\ell}\right], \tag{5.23}$$

where $z_+ = 0$ and $z_- = d$. Averaging $g_\pm$ over z, we have

$$g_\pm(t) = \frac{e\tau E}{m}\frac{\partial f_0}{\partial v_\parallel}\left[1 - \frac{\ell}{td}\frac{[1-p(t)](1-e^{-td/\ell})}{1-p(t)e^{-td/\ell}}\right]. \tag{5.24}$$

Let us consider first the case of $p(t) = 1$. In this case $g_\pm$ are uniform and the conductivity is identical to the bulk conductivity given by Eq. (3.8). In the case of $p(t) < 1$, $g_\pm$ will have a dependence on the momentum of the electrons. Using $k_\parallel = k_F\sqrt{1-1/t^2}$ and $\frac{\partial f_0}{\partial k_\parallel} = \delta(k-k_F)k_\parallel/k$, we obtain the conductivity as

$$\sigma_{xx} = \sigma_0\left\{1 - \frac{3\ell}{2d}\int_1^\infty dt\left(\frac{1}{t^3} - \frac{1}{t^5}\right) \times \frac{[1-p(t)](1-e^{-td/\ell})}{1-p(t)e^{-td/\ell}}\right\}, \tag{5.25}$$

where σ_0 is the bulk conductivity given by Eq. (3.8). The integral over t can be evaluated exactly in the limit of $\ell \to \infty$. In this limit, we replace the exponential function $e^{-td/\ell}$ by a rational function,

$$1 - \frac{td/\ell}{1+td/2\ell}, \tag{5.26}$$

which has the same value and the first two derivatives as the original function at the limit $td/\ell \to 0$. Thus assuming that $p(t)$ is constant, the conductivity in the limit of large mean free path is

$$\sigma_{xx} = \frac{e^2k_F^2d}{12\pi^2\hbar}\frac{(1+p)}{(1-p)}\ln\frac{\ell}{d}. \tag{5.27}$$

Comparing this result with the logarithmic form derived from the Kubo formula, Eq. (5.17), we can relate ξ_1, which correlates the broadening of the bands with the z component of the momentum, to the macroscopic parameter p by

$$\xi_1 = \frac{3}{d}\frac{(1-p)}{(1+p)}. \tag{5.28}$$

The Fuchs-Sondheimer assumption that p is a constant agrees in the limit $\ell/d \to \infty$ with an angle dependence of the mean free path of the form $\ell(t) = \ell_0/(1+\xi_1\ell_0\cos\theta)$, which we have shown to correspond to a nonanalytic self-energy. Using a $p \neq 1$ when $\theta = 90°$ is clearly unphysical because when the z component of the velocity is zero, we should have $g^+ = g^-$ which implies $p(t) \to 1$. As the angle of incidence approaches the glancing angle the electron travels longer before it hits the surface and therefore its mean free path should approach that of the bulk continuously. In fact, there is a one to one relationship between $\ell(t)$ and $p(t)$, which can be obtained by requiring that the semiclassical expression for the conductivity involving $\ell(t)$, Eq. (5.10) agree with Eq. (5.25). This requirement implies

$$p(t) = 1 - \frac{t}{\frac{\ell_0\ell(t)}{[\ell_0-\ell(t)]d} - \frac{te^{-td/\ell(t)}}{1-e^{-td/\ell(t)}}}. \tag{5.29}$$

If we use the relationship Eq. (5.16) then we have

$$p(t) = 1 - \frac{\xi_1 d}{1 - \frac{\xi_1 d}{e^{td/\ell_0+\xi_1 d}-1}}. \tag{5.30}$$

In the limit of $t \to \infty$ (i.e., $\theta \to 90°$), we have $p(t) \to 1-\xi_1 d$. This shows that any value of $p \neq 1$ at the glancing angle corresponds to the form given by Eq. (5.16) at least for θ close to 90°. As we have noted, this introduces a nonanalyticity of the self-energy at $k_z = 0$. To correct this we use the form predicted by the quantum theory, Eq. (5.12). Substituting that equation into Eq. (5.29), we get

$$p(\cos\theta) = 1 - \frac{1}{\frac{1}{\xi_2 k_F a\cos\theta} - \frac{1}{e^{\frac{d}{\ell_0\cos\theta}+\xi_2 k_F a\cos\theta}-1}}. \tag{5.31}$$

Now we have properly $p(t) \to 1$ in the limit of $\theta \to 90°$.

D. Comparison with experiment

In Fig. 4 the predictions of three theories based on the FERPS model are compared with the experimental resistivities of Sn, Pt, and $CoSi_2$ thin films. The solid lines represent fits to the continuum limit of the quantum theory as given by Eq. (5.13), which uses the correct angular dependence of the lifetime. The dashed lines also represent fits to the continuum limit of the quantum theory, but as given by Eq. (5.17) which assumes an angular dependence of the mean free path as given by Eq. (5.16). The dotted lines are fits to Fuchs-Sondheimer theory with $p = 0$.

There are two independent fitting parameters for the fits using Eqs. (5.13) and (5.17), k_F and $\xi_2\ell_0$ or $\xi_1\ell_0$. The same value of k_F was used for both versions of the quantum theory. Note that Fig. 4(c) also shows the experimental resistivity for a silicon-capped $CoSi_2$ film, which has very little surface scattering. This data is fit using Eq. (5.13) and the same k_F as for the uncapped $CoSi_2$ film. For the fits using Fuchs-Sondheimer theory there were also, in principle, two parameters, ℓ and p, but $p = 0$ always gave the best fit.

We note that Fuchs-Sondheimer theory can be fit to the experimental data only for relatively thick films. Significant deviations from the experiments occur for film thicknesses less than a few hundred Angstroms. This deviation comes mainly from two parts, the angular depen-

dence of the mean free path (and consequently of p), and the quantum cutoff of the z component of the momentum k_z at π/d (effectively the zero-point motion in the z direction). Both effects are important, as we could not fit the Pt and $CoSi_2$ experiments using Fuchs-Sondheimer theory with a constant p even when the integration over t is cut off at $k_F d/\pi$.

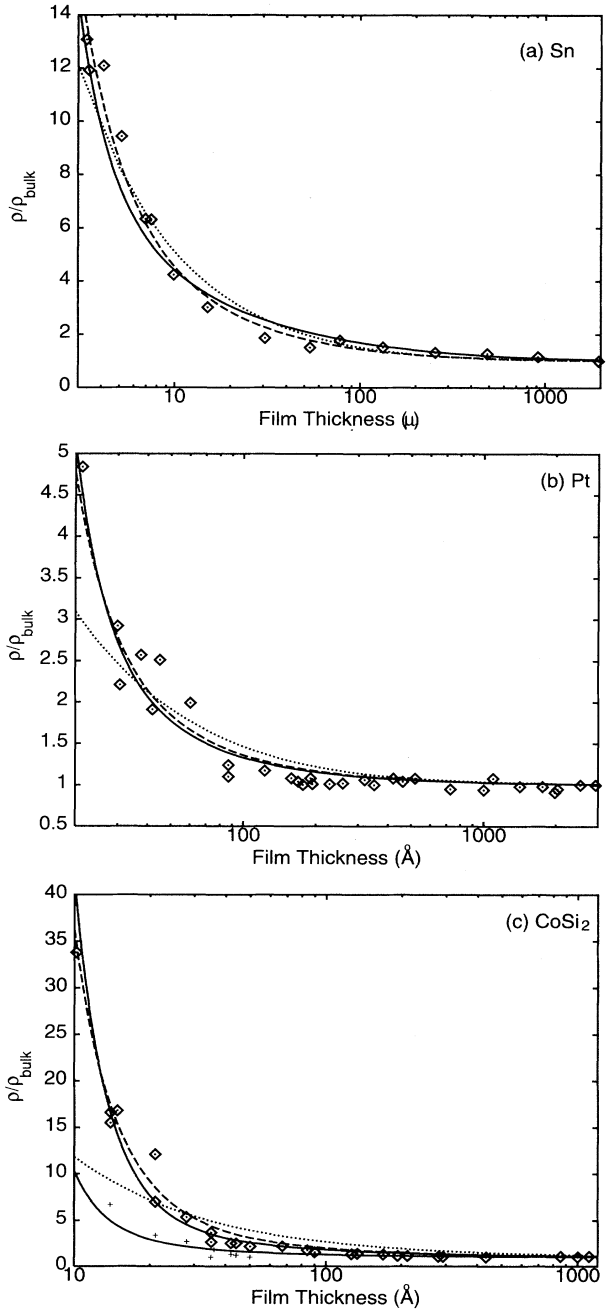

FIG. 4. Resistivity as a function of film thickness for (a) foiled Sn (Ref. 22), (b) Pt films (Ref. 23), and (c) epitaxial $CoSi_2$ films (Refs. 24 and 27). Solid lines are predictions of Eq. (5.13), dashed lines are the predictions of Eq. (5.17), and the dotted lines are the predictions of Fuch-Sondheimer theory with $p = 0$. Capped $CoSi_2$ data are shown as crosses.

The continuum limit of the quantum theory, which includes the electronic zero-point motion gives excellent fits to these experiments down to the thinnest films, even when other quantum effects are neglected. This is true both for the form of Eq. (5.13) and for the form of Eq. (5.17). From these experimental data alone one cannot determine which form, Eq. (5.13) or Eq. (5.17), is superior although we prefer the former for reasons already given.

VI. MULTILAYERS

In this section, we present numerical calculations of the conductivity for multilayers within the FERPS model and compare these results with those of the semiclassical theories and the approximate quantum theory of Zhang, Levy, and Fert. Although the quantum theory for thin films presented in the previous section could be applied to treat multilayers or superlattices, that approach does not allow for the wave functions to be modified by inhomgeneities in the scattering rate so it would be limited to cases in which the layer thicknesses are less than the mean free path or to cases in which the mean free path is approximately the same for all layers.

In order to apply the Kubo-Greenwood linear response theory to layers of arbitrary thickness we numerically calculate the partial Green function, $G(k_\parallel; z, z')$, which satisfies Eq. (5.1), by using the general solution,[15]

$$G(k_\parallel; z, z') = \frac{\psi_L(z_<)\psi_R(z_>)}{W}, \tag{6.1}$$

where $\psi_L(z)$ and $\psi_R(z)$ are solutions to the homogeneous part of Eq. (5.1), which satisfy boundary conditions on the left and right sides, respectively, of the multilayer, and where W is the Wronskian of ψ_L and ψ_R. For a multilayer system of total thickness d we used boundary conditions, $\psi_L(0) = 0$ and $\psi_R(d)=0$. After the partial Green function was obtained, the conductivity was calculated from Eq. (5.4).

The conductivity for a series of superlattices is shown in Fig. 5 and compared with approximate semiclassical and quantum results. In these calculations it is assumed that the scattering rates for the two layers correspond to bulk mean free paths of 36.0555 and 360.555 a.u. and that the thickness of the dirty layers is twice that of the clean layers. No additional scattering at the interfaces is included. In all of the multilayer calculations, we used a sufficient number of periods of the multilayer to avoid the physical quantum size effects for the exact results and the unphysical size effects that occur for the semiclassical and ZLF theories.

Figure 5 also shows the conductivity in the thin and the thick limits. In the thick limit ($d \gg \ell_{\text{eff}}$) one simply adds conductivities in parallel so that the conductivity is proportional to an effective mean free path, which is just the average of the mean free paths in the separate regions,

$$\ell_{\text{thick}} = \sum_I \frac{d_I}{d} \ell_I. \tag{6.2}$$

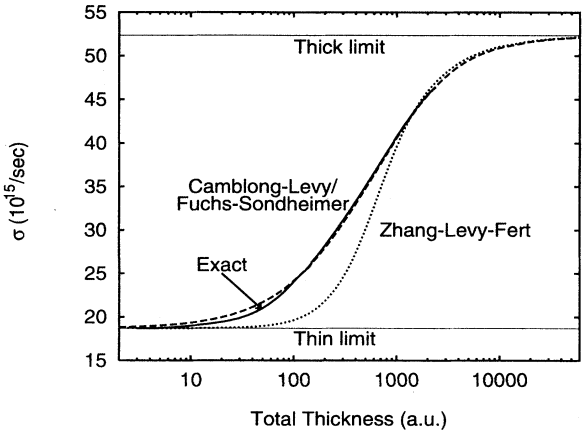

FIG. 5. The conductivity as a function of the total thickness of a period for a periodic multilayer with a period of two layers and with mean free paths in each layer 36.0555 a.u. and 360.555 a.u., respectively. (1 a.u. = 0.529 Å.) The thickness of the first layer is twice that of the second. In this case Camblong and Levy's theory is identical to the Fuchs-Sondheimer theory with $p = 1$.

In the thin limit ($d \ll \ell_{\rm eff}$), we can use the golden rule to show that the mean free path of electrons in the multilayer system is given by

$$\frac{1}{\ell_{\rm thin}} = \sum_I \frac{d_I}{d}\frac{1}{\ell_I} + \text{oscillatory terms.} \tag{6.3}$$

This means that on the length scale of the mean free path, the in-plane conductivity is given by the average of the inverse mean free path. Note that in the case of surface (or interface) roughness, where a thin region contains very strong scattering described by a very short effective bulk mean free path, it will contribute significantly to the mean free path of the system.

The Zhang-Levy-Fert approximate evaluation of the Kubo formula is given by Eqs. (2.19), (2.20), and (2.21). Note that their method cannot be applied to films, because it does not include finite size effects. In fact a size effect with the wrong sign would arise for a film, i.e., the conductivity would increase as the thickness of the film decreased, if one were to apply their theory to a film of finite thickness. The Camblong-Levy approximation to the conductivity of the multilayer can be obtained by integrating Eq. (2.17) over z' and z,

$$\sigma^{II} = \frac{c_D}{d_I}\left\{\ell_I d_I + \frac{3}{4}\ell_I^2\left[-\frac{1}{2} + 2E_3\left(\frac{d_I}{\ell_I}\right) - 2E_5\left(\frac{d_I}{\ell_I}\right)\right]\right\}, \tag{6.4}$$

and

$$\sigma^{IJ} = \frac{3c_D}{4d_I}\ell_I\ell_J[E_3(\phi_{IJ}) - E_3(\phi_{I-1,J}) - E_3(\phi_{I,J+1}) + E_3(\phi_{I-1,J+1}) - E_5(\phi_{IJ}) + E_5(\phi_{I-1,J}) + E_5(\phi_{I,J+1}) - E_5(\phi_{I-1,J+1})], \tag{6.5}$$

where $c_D = e^2k_F^2/6\pi^2\hbar$, and the ϕ_{IJ} are defined by

$$\phi_{IJ} = \int_{z_{I+1}}^{z_J}\frac{dz}{\ell(z)} \qquad (I < J). \tag{6.6}$$

The above Camblong-Levy expressions for the conductivity can also be derived from the Fuchs-Sondheimer theory using a z-dependent lifetime $\tau(z)$, or τ_I for layer I in the multilayer case. It is straightforward to show that in this case the distribution function $g^{IJ}(\mathbf{v})$ as given by Eqs. (B9) and (B10) becomes

$$g^{IJ}(\mathbf{v}) = \frac{e\tau_I\tau_J v_z}{md_I}\boldsymbol{\mathcal{E}}_J\cdot\nabla_{\mathbf{v}}f_0(1-e^{-\frac{d_I}{\tau_I v_z}}) \times(1-e^{-\frac{d_J}{\tau_J v_z}})e^{-\phi_{IJ}\frac{v}{v_z}}, \quad I > J, \tag{6.7}$$

$$g^{II}(\mathbf{v}) = \frac{e\tau_I}{m}\boldsymbol{\mathcal{E}}_I\cdot\nabla_{\mathbf{v}}f_0\left[1-\frac{\tau_I v_z}{d_I}(1-e^{-\frac{d_I}{\tau_I v_z}})\right]. \tag{6.8}$$

These equations in turn lead to the conductivity as given by Eq. (6.5). The truncation of the sum over σ^{IJ} in the case of a film with finite thickness is equivalent to complete diffuse surface scattering, which would be modeled in Fuchs-Sondheimer theory by $p = 0$ in the boundary conditions at the surfaces.

From Fig. 5, it is clear that although the ZLF theory is correct in both the limits of very thin films and very thick films, it does not accurately reproduce the exact results for intermediate film thicknesses. In particular, the ZLF conductivity remains very close to the thin limit until the total film thickness reaches about 100 a.u. This can be understood from the ZLF equations. Since the scattering rate in the thicker dirty layers is ten times higher than in the cleaner thin layers and the dirty layers are twice the thickness of the clean layers, the average mean free path of Eq. (2.21) is approximately 1.5 times the mean free path of the dirty layer. Thus, the scattering rate which enters the ZLF conductivity is averaged over a thickness of approximately 54 a.u. The ZLF theory will only depart from the thin limit when the thickness of the dirty layers exceeds this thickness, i.e., when the total thickness of a multilayer period is about 81 a.u.

The conductivity calculated from the semiclassical theory agrees surprisingly well with the numerical evaluation of the Kubo formula. Small differences can be observed in Fig. 5 in the regime in which the thickness of the dirty layer is comparable to the mean free path of that layer. More insight can be gained from inspection of the layer dependent conductivity, $\sigma(z)$, which is shown in Fig. 6 for three thicknesses of the multilayer period. Figure 6(a) shows $\sigma(z)$ for a multilayer period of 30 a.u. The clean layer extends from 10 to 20 a.u. This multilayer is in the thin limit because both layer thicknesses are less than the shortest mean free path (36.0555 a.u.). The conductivity in the ZLF approximation rises from approximately 17×10^{15}/sec, to approximately 21×10^{15}/sec, whereas the semiclassical theory has a much larger rise. The increase of the exact $\sigma(z)$ in the clean layer is intermediate.

Figure 6(b) shows $\sigma(z)$ for a longer multilayer period. The dirty layers are 200 a.u. thick and the clean layers are 100 a.u. in thickness and extend from 100 a.u. to 200 a.u. on the figure. The exact and semiclassical results are now close, but the ZLF conductivity is significantly

lower in the clean layer. Figure 6(c) shows $\sigma(z)$ for a multilayer period of 900 a.u. The dirty layers are 600 a.u. thick and the clean layers are 300 a.u. in thickness. The semiclassical $\sigma(z)$ is now in very good agreement with that obtained from the numerical evaluation of the Kubo formula. We intentionally performed the numerical calculations for the quantum solution using a single period in this thick multilayer in order to display the scale of the quantum size effect, which can be seen as a small spike and oscillations at the edges of the film. We believe that the very small oscillations seen elsewhere in the calculated results are due to incomplete convergence of the integral over $k_{\|}$.

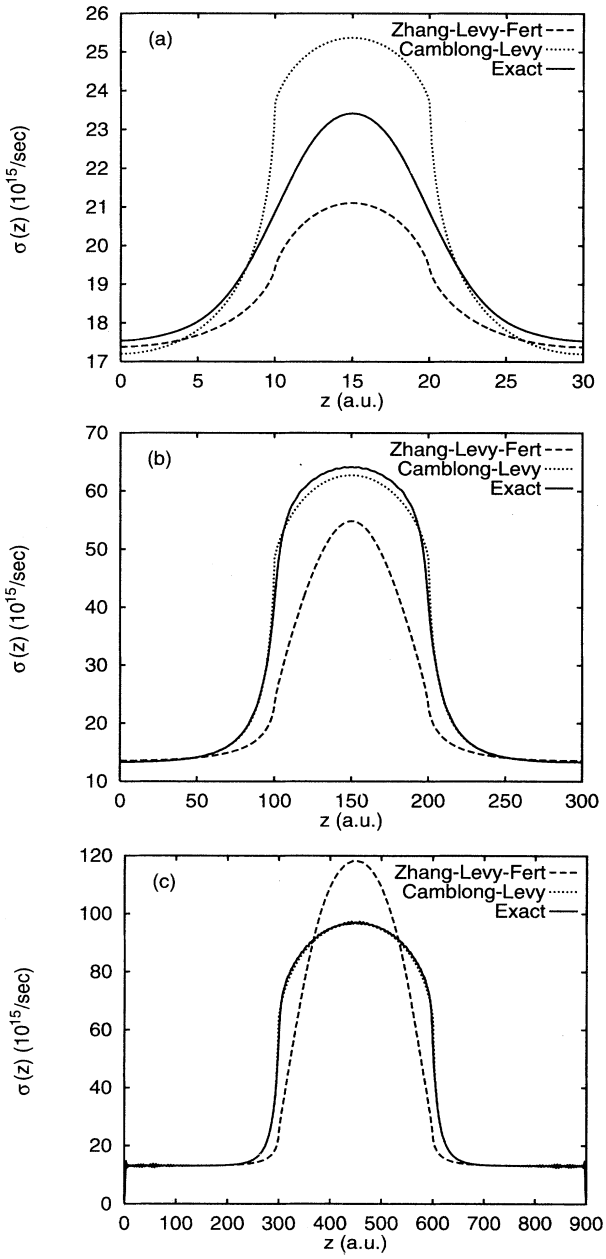

FIG. 6. Conductivity as a function of z, $\sigma_{xx}(z)$ for multilayers. The central clean layer has a mean free path of 360.555 a.u. and one-half the thickness of the dirty layer, which has a mean free path of 36.0555 a.u. (a) Period of multilayer is 30 a.u.; clean layer lies between 10 and 20 a.u. (b) Period of multilayer is 300 a.u.; clean layer lies between 100 and 200 a.u. (c) Period of multilayer is 900 a.u.; clean layer lies between 300 and 600 a.u. (1 a.u. = 0.529 Å.)

These figures illustrate one problem with the ZLF approximation. The average mean free path calculated from Eq. (2.21) is the same for all of the multilayer systems shown here because the ratio of the thicknesses of the clean and dirty layers is fixed at 1/2. Thus, from Eq. (2.20) we see that inverse lifetime is averaged over the same distance at each point for all of these multilayers. This does not seem correct to us. Intuitively one would expect that at the center of the dirty layer the effective mean free path for averaging the scattering rate should be that of the dirty layer if this layer is sufficiently thick. Similarly, one would expect that at the center of a sufficiently thick clean layer, the effective mean free path for averaging the scattering rate should be that of the clean layer not an average that is dominated by the strong scattering of the dirty layer.

Finally, in Fig. 7 we show the calculated GMR effect for a multilayer. The conductivity was calculated for a model system consisting of a central spacer layer with a fixed thickness of 50 a.u. surrounded by strongly scattering interfacial layers 4 a.u. in thickness (approximately one atomic layer). These are surrounded by magnetic layers of variable thickness. The mean free path for both spins was taken to be 427 a.u. in the spacer layer. The mean free paths were assumed to be 50 a.u. and 10 a.u., respectively, for the majority and minority electrons in the thin interfacial layers and 100 a.u. and 20 a.u. for majority and minority electrons, respectively, in the ferromagnetic layers.

Panel (a) shows the calculated conductivity due to the majority electrons in the parallel moment configuration as given by the exact quantum result as well as the semiclassical theory and the approximate quantum theory of ZLF; panel (b) shows the calculated conductivity due to the minority spin electrons in the parallel moment configuration; panel (c) shows the conductivity for either spin channel in the antiparallel alignment; and panel (d) shows the difference between the conductivities of the parallel and antiparallel configurations divided by the conductivity of the parallel alignment. The ZLF conductivities are all lower than the exact or semiclassical conductivities and the giant magnetoresistance, $\Delta\sigma/\sigma_{\|}$, is much larger. The semiclassical conductivities and GMR are much closer to the exact ones, but the differences are still significant.

VII. CONCLUSION

We have solved the Kubo formula exactly for the free-electron model with random point scattering for the cases of a homogeneous system, a thin film, and a multilayer. Using the exact solutions we compared various semiclassical approaches as well as an approximate solution of the

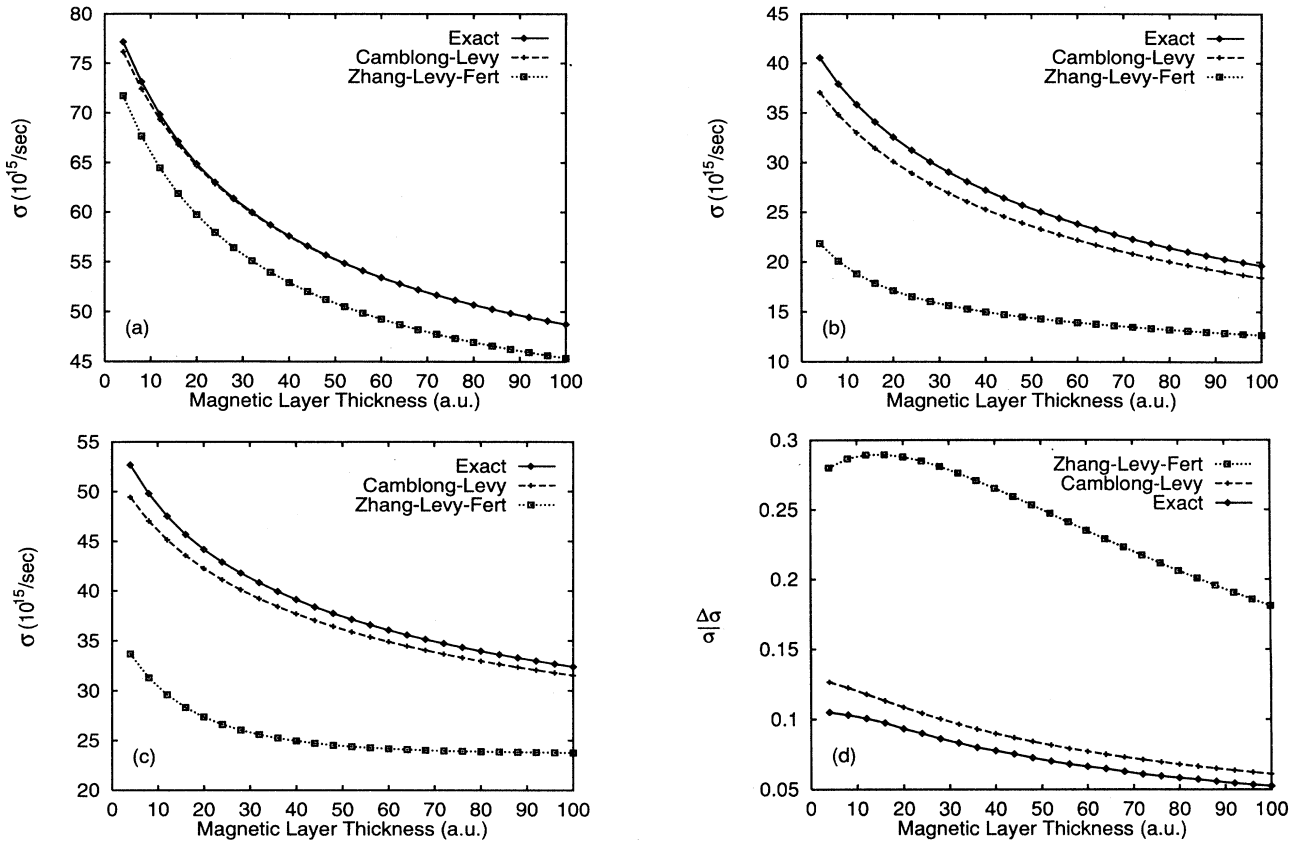

FIG. 7. Calculated conductivity of a model GMR multilayer system as a function of the thickness of the magnetic layer. The spacer layer thickness is fixed at 50 a.u. and has a mean free path of 427 a.u. There are thin interfacial layers (4 a.u.) separating the spacer and ferromagnetic layers. (a) Conductivity in the majority channel with magnetic layers parallel. The mean free path in the ferromagnetic layers is 100 a.u. and in the interfacial layer is 50 a.u. (b) Conductivity in the minority channel with magnetic layers parallel. The mean free path in the ferromagnetic layer is 20 a.u. and in the interfacial layers is 10 a.u. (c) Conductivity in either channel with magnetic layers antiparallel. (d) Difference between parallel and antiparallel conductivities divided by parallel conductivity. (1 a.u. = 0.529 Å.)

Kubo formula. For the case of a homogeneous system we show that all semiclassical approaches are equivalent, and give reasonable approximations of the two-point conductivity compared to the quantum solution. For the case of thin films, we show that there are two important differences between our results and Fuchs-Sondheimer theory, a quantum effect resulting from the cutoff of the momentum in the z direction (an effective zero-point motion of the electrons), and the angular dependence of the mean free path, or in the case of Fuchs-Sondheimer theory the angular dependence of the roughness parameter p. When both effects are included these theories give excellent agreement with experimental data on several thin films. Finally in the case of multilayers, we found that all of these approaches give the correct thin and thick limits, but the ZLF approach does not work as well for intermediate thicknesses as the semiclassical approaches. The effect of these errors on the GMR can be quite large.

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APPENDIX A: LAYER CONDUCTIVITY

Here, we derive Eqs. (3.10) and (3.11) starting from Eq. (3.9). Since the system is homogeneous, we can write

$$\sigma_{zz}(Z) = \frac{e^2Z^2}{4\pi^3\hbar}\int dX \int dY \left\{ \mathrm{Im}\left[e^{i\kappa R}\left(i\kappa - \frac{1}{R}\right)\right]\right\}^2 \frac{1}{R^4}, \tag{A1}$$

where $X = x - x'$, $Y = y - y'$, $Z = z - z'$, and $R^2 = X^2 + Y^2 + Z^2$. Using $\rho^2 = X^2 + Y^2$ and

$$\int dX \int dY = 2\pi \int_0^\infty \rho d\rho = 2\pi \int_Z^\infty R dR, \qquad \text{(A2)}$$

we obtain

$$\sigma_{zz}(Z) = \frac{e^2 Z^2}{8\pi^2 \hbar} \int_Z^\infty \frac{dR}{R^3} \left[e^{i\kappa R} \left(i\kappa - \frac{1}{R} \right) - e^{-i\kappa^* R} \left(-i\kappa^* - \frac{1}{R} \right) \right]^2 . \qquad \text{(A3)}$$

Making a change of variables $R \to Zt$ and using the definition of the exponential integral, Eq. (3.12), we obtain Eq. (3.10).

Similarly, $\sigma_{xx}(Z)$ can be derived using

$$\sigma_{xx}(Z) = \frac{1}{2}[\sigma_{xx}(Z) + \sigma_{yy}(Z)], \qquad \text{(A4)}$$

$$\sigma_{xx}(Z) = \frac{e^2}{16\pi^2 \hbar} \int_Z^\infty \frac{dR}{R^3}(R^2 - Z^2) \left[e^{i\kappa R} \left(i\kappa - \frac{1}{R} \right) - e^{-i\kappa^* R} \left(-i\kappa^* - \frac{1}{R} \right) \right]^2 . \qquad \text{(A5)}$$

With the same change of variable used above, we obtain Eq. (3.11).

APPENDIX B: TWO-POINT CONDUCTIVITY OF FUCHS-SONDHEIMER THEORY FOR MULTILAYERS

The g^{IJ} as defined in Eq. (4.25) satisfy the differential equations,

$$\left[\mathbf{v} \cdot \nabla + \frac{1}{\tau} \right] g^{IJ}(\mathbf{v}, z) = \frac{e}{m} \delta_{IJ} \boldsymbol{\mathcal{E}}_J \cdot \nabla_{\mathbf{v}} f_0. \qquad \text{(B1)}$$

Since $\mathcal{E}_J$ is confined within a layer J, the boundary conditions at $z = \pm\infty$ is $g^{IJ} \to 0$, for $I \to \pm\infty$. If v_z is greater than zero, g^{IJ} will be nonzero only for $I \geq J$, similarly for v_z less than zero, g^{IJ} will be nonzero for $I \leq J$. Thus for $v_z > 0$ we have, in order to satisfy Eq. (B1),

$$g_+^{IJ}(\mathbf{v}, z) = 0, \quad I < J, \qquad \text{(B2)}$$

$$g_+^{JJ}(\mathbf{v}, z) = \frac{e\tau}{m} \boldsymbol{\mathcal{E}}_J \cdot \nabla_{\mathbf{v}} f_0 [1 + F_+^{JJ}(\mathbf{v}) e^{-\frac{z}{\tau v_z}}], \qquad \text{(B3)}$$

$$g_+^{IJ}(\mathbf{v}, z) = \frac{e\tau}{m} \boldsymbol{\mathcal{E}}_J \cdot \nabla_{\mathbf{v}} f_0 F_+^{IJ}(\mathbf{v}) e^{-\frac{z}{\tau v_z}}, \quad I > J. \qquad \text{(B4)}$$

The subscript + indicates the direction of v_z. Since the $g_+^{IJ}(\mathbf{v}, z)$ must be continuous at the interfaces, we have for z on the interface between the layers I and $I + 1$,

$$g_+^{I+1,J}(\mathbf{v}, z) = g_+^{IJ}(\mathbf{v}, z). \qquad \text{(B5)}$$

This condition leads to

$$F_+^{I+1,J}(\mathbf{v}) = F_+^{IJ}(\mathbf{v}) \quad \text{for } I > J,$$
$$F_+^{J+1,J}(\mathbf{v}) = e^{\frac{d}{\tau v_z}} + F_+^{JJ}(\mathbf{v}),$$
$$F_+^{JJ}(\mathbf{v}) + 1 = 0. \qquad \text{(B6)}$$

Therefore, we find the solutions for g_+^{IJ} ($v_z > 0$),

$$g_+^{IJ}(\mathbf{v}, z) = \frac{e\tau}{m} \boldsymbol{\mathcal{E}}_J \cdot \nabla_{\mathbf{v}} f_0 [1 - e^{-\frac{d}{\tau v_z}}] e^{-\frac{z-d}{\tau v_z}}, \quad I > J, \qquad \text{(B7)}$$

$$g_+^{II}(\mathbf{v}, z) = \frac{e\tau}{m} \boldsymbol{\mathcal{E}}_J \cdot \nabla_{\mathbf{v}} f_0 [1 - e^{-\frac{z}{\tau v_z}}]. \qquad \text{(B8)}$$

We further average $g_+^{IJ}(\mathbf{v}, z)$ over the layer I, and obtain

$$g_+^{IJ}(\mathbf{v}) = \frac{e\tau^2 v_z}{md} \boldsymbol{\mathcal{E}}_J \cdot \nabla_{\mathbf{v}} f_0 (1 - e^{-\frac{d}{\tau v_z}})^2 e^{-\frac{(I-J-1)d}{\tau v_z}}, \quad I > J, \qquad \text{(B9)}$$

$$g_+^{II}(\mathbf{v}) = \frac{e\tau}{m} \boldsymbol{\mathcal{E}}_I \cdot \nabla_{\mathbf{v}} f_0 \left[1 - \frac{\tau v_z}{d} (1 - e^{-\frac{d}{\tau v_z}}) \right]. \qquad \text{(B10)}$$

g_-^{IJ} can be found by a space inversion, $z \to -z$, which leads to

$$g_-^{IJ}(\mathbf{v}) = g_+^{-I,-J}(-\mathbf{v}). \qquad \text{(B11)}$$

Using Eq. (4.26), we can obtain (for E in the z direction)

$$\sigma_{zz}^{IJ} = \frac{1}{(2\pi)^2} \frac{e\hbar}{m} \int k^3 dk \left[\int_{-1}^0 d\cos\theta \cos\theta g_-^{IJ} + \int_0^1 d\cos\theta \cos\theta g_+^{IJ} \right]. \qquad \text{(B12)}$$

If we use $\partial f_0 / \partial v_z = m v_z \delta(\epsilon - \epsilon_F)$, $v_z = v\cos\theta$ and make a change of variables $\cos\theta = 1/t$, we obtain Eqs. (4.27) and (4.28).

Similarly, for E in the x direction,

$$\sigma_{xx}^{IJ} = \frac{1}{(2\pi)^2} \frac{e\hbar}{m} \int k^3 dk \int_0^{2\pi} d\phi \cos\phi \left[\int_{-1}^0 d\cos\theta \sin\theta g_-^{IJ} + \int_0^1 d\cos\theta \sin\theta g_+^{IJ} \right]. \qquad \text{(B13)}$$

Using $v_x = v\sin\theta\cos\phi$ and $\partial f_0 / \partial v_x = 2m v_x \delta(\epsilon - \epsilon_F)$, we obtain the conductivity for the current in-plane (CIP) case, Eqs. (4.29) and (4.30).

APPENDIX C: SOLUTION FOR THIN FILM

We outline here the derivation for the thin film conductivity. The Kubo formula for a film, Eq. (5.4), can be written as

$$\sigma_{\parallel} = -\frac{1}{8\pi^3}\frac{e^2\hbar^3}{m^2 d}\int_0^d dz\int_0^d dz'\int d^2k_{\parallel}k_{\parallel}^2\frac{1}{4}[G\bar{G}+G^*\bar{G}^* - G\bar{G}^* - G^*\bar{G}] \tag{C1}$$

where the Green functions, $G = G(k_{\parallel}; z, z')$ and $\bar{G} = G(k_{\parallel}; z', z)$ are given by Eq. (5.2). Using the fact that $G(k_{\parallel}; z, z') = G(k_{\parallel}; z', z)$, which is valid in the absence of effects such as spin orbit coupling which break time reversal invariance for a given spin, we can perform the integrals over z and z' by using the normalization conditions for the one-dimensional wave functions $\phi_n(z)$,

$$\int_0^d dz\phi_m^*(z)\phi_n(z) = \delta_{mn}. \tag{C2}$$

Then the expression for σ simplifies to

$$\sigma_{\parallel} = -\frac{1}{8\pi^3}\frac{e^2\hbar^3}{m^2 d}\int d^2k_{\parallel}k_{\parallel}^2\sum_n I_n^2, \tag{C3}$$

where

$$I_n = \mathrm{Im}\frac{1}{E-\Sigma-\epsilon_n-\dfrac{\hbar^2k_{\parallel}^2}{2m}}. \tag{C4}$$

Now the integration over $\mathbf{k}_{\parallel}$ can be carried out,

$$\int d^2\mathbf{k}_{\parallel}k_{\parallel}^2 I_n^2 = -\frac{\pi}{2}\int_0^{\infty} dk_{\parallel}k_{\parallel}^3\left(\frac{1}{E-\Sigma-\epsilon_n-\dfrac{\hbar^2k_{\parallel}^2}{2m}} - \frac{1}{E-\Sigma^*-\epsilon_n-\dfrac{\hbar^2k_{\parallel}^2}{2m}}\right)^2. \tag{C5}$$

Defining $u = k_{\parallel}^2$, $\alpha_n = 2m(E-\epsilon_n)/\hbar^2$, and $\eta = 2mi\Sigma/\hbar^2$, we have

$$\int d^2\mathbf{k}_{\parallel}k_{\parallel}^2 I_n^2 = -\frac{\pi m^2}{\hbar^4}\int_0^{\infty} udu\left(\frac{1}{\alpha_n+i\eta-u} - \frac{1}{\alpha_n-i\eta-u}\right)^2, \tag{C6}$$

which can be integrated to yield,

$$\int d^2\mathbf{k}_{\parallel}k_{\parallel}^2 I_n^2 = \begin{cases} \dfrac{2\pi m^2}{\hbar^4}\left[1-\dfrac{\alpha_n}{\eta}\arctan\dfrac{\eta}{\alpha_n}\right], & \alpha_n < 0 \\ \dfrac{2\pi m^2}{\hbar^4}\left[1+\dfrac{\alpha_n}{\eta}\left(\pi-\arctan\dfrac{\eta}{\alpha_n}\right)\right], & \alpha_n \geq 0. \end{cases} \tag{C7}$$

Substituting this back into Eq. (C3) yields the result given by Eq. (5.5).

APPENDIX D: LIFETIME OF ELECTRONS IN A THIN FILM

If the elementary scattering centers are distributed nonuniformly within the film the imaginary part of the self-energy [η in Eq. (5.5)] will depend on n. To lowest nontrivial order, this scattering rate can be obtained from the golden rule. It can also be obtained by more sophisticated techniques.[28] We distribute point scatterers of strengths $V_i\delta(\mathbf{r}-\mathbf{r}_i)$ with a density $c(z)$ randomly throughout the film. For simplicity, and without losing generality, we assume that all scatterers have the same magnitude of scattering strength but their signs are randomly distributed. According to the golden rule, the probability of an electron scattering from subband n to subband n' is

$$P_{nn'} = \frac{2\pi}{\hbar}\sum_{k_{\parallel}'}|\langle\psi_n(\mathbf{k}_{\parallel})|\Delta H|\psi_{n'}(\mathbf{k}_{\parallel}')\rangle|^2\delta(E-E'). \tag{D1}$$

The wave functions in x and y directions are described by plane waves so the integrals over these directions in Eq. (D1) can be performed giving

$$P_{nn'} = \frac{mV^2}{\hbar^3}\int_0^d dzc(z)|\phi_n(z)\phi_{n'}(z)|^2, \tag{D2}$$

where we have assumed that $V = |V_i|$ and V_i averages to zero. The lifetime of an electron in subband n can be found from

$$\frac{1}{\tau_n} = \sum_{n'=1}^{N} P_{nn'}, \tag{D3}$$

where $N = k_F d/\pi$. Using the wave function for an infinite square well, $\phi_n(z) = \sqrt{2/d}\sin n\pi z/d$, we have

$$\frac{1}{\tau_n} = \frac{mV^2}{d^2\hbar^3}\int_0^d dzc(z)\left[2N+1-\frac{\sin(2N+1)\dfrac{\pi z}{d}}{\sin\dfrac{\pi z}{d}} - 4\sin^2\frac{n\pi z}{d}\right]\sin^2\frac{n\pi z}{d}. \tag{D4}$$

For a uniform distribution of scatterers $c(z)$ is a constant and the lifetime is independent of the energy levels,

$$\frac{1}{\tau_n} = \frac{1}{\tau_0} \equiv \frac{mV^2c(N-1)}{\hbar^3 d} = \frac{mV^2c(k_F-\pi/d)}{\pi\hbar^3}. \tag{D5}$$

If there are extra scatterers, Δc_i, distributed in a region around $z = z_i$ of thickness Δd, then

$$\frac{1}{\tau_n} = \frac{1}{\tau_0}\left\{1 + \frac{\Delta d}{d}\frac{\Delta c_i}{c(N-1)}\left[2N+1 - \frac{\sin(2N+1)\frac{\pi z_i}{d}}{\sin\frac{\pi z_i}{d}} - 4\sin^2\frac{n\pi z_i}{d}\right]\sin^2\frac{n\pi z_i}{d}\right\}. \quad \text{(D6)}$$

In most cases the extra scattering is near the surfaces, so we can expand the above equation in the distances to the surfaces, and get Eq. (5.6). Even when the expansion of the above equation in a Taylor series of z_i is invalid, it is easy to see that the lifetime is an even function of n, or $k_z = n\pi/d$, and Eq. (5.6) can still be a reasonable approximation.

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