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# Transport theory in metallic films: Crossover from the classical to the quantum regime

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Using the quantum-statistical approach, we have developed a unified transport theory of metallic films. A general description for the conductivity in metallic films has been rigorously formulated in the presence of both impurity scattering and surface roughness. An explicit connection between the quasiclassical and present quantum approaches is also presented. We show that the quasiclassical theory by Fuchs [Proc. Cambridge Philos. Soc. 34, 100 (1938)] and Sondheimer [Adv. Phys. 1, 1 (1952)] can be reformed to be applicable to ultrathin metallic films by introducing a treatment of the surface via angle-dependent specularity parameters and including the quantum size effect. Moreover, to the lowest order approximation in the theory, the previous quantum-approach results and discussions have naturally been recovered.

Following advances in microfabrication of nanostructures such as metallic multilayers<sup>1</sup> there has been a revival of research activities on transport properties of metallic films. Since a thin film is a basic unit of a metallic multilayer, theoretical approaches to the transport in metallic films<sup>2-6</sup> have been widely applied to metallic multilayers. 7-10 Besides, investigations in connection with the metallic films themselves have also been interesting and important. There have been two conceptually different approaches to the transport in metallic films: quasiclassical and quantum. The quasiclassical theory developed by Fuchs and Sondheimer<sup>2</sup> is based on the Boltzmann equation in the presence of a diffusely reflecting boundary. The surface scattering is characterized by a specularity parameter R according to the surface roughness, with R lying between 0 (for completely diffusive scattering) and 1 (for completely specular reflection). It is well known that the quasiclassical approach is inadequate for very thin, high-purity samples where the film thickness d is much smaller than the impurity mean free path  $\lambda$ , although it works well for thick, relatively dirty systems. In particular, in the limit when  $\lambda$  goes to infinity the conductivity diverges as  $\ln(\lambda/d)$ . This nonphysical result arises from a complete omission of quantum size effects from the quasiclassical theory. In order to overcome these shortcomings and study the quantum effects in metallic films, several quantum surface scattering treatments have been proposed by Tešanović et al.,4 Trivedi and Ashcroft (TA),<sup>5</sup> and Fishman and Calecki (FC). These quantum-mechanism approaches are based on the Kubo linear response theory in which the surface roughness is incorporated into the Hamiltonian. They focus attention on the quantum size effects describing the increase in resistivity of a film as d becomes shorter than  $\lambda$  and make a success of accounting for experimental data on thin metallic films of CoSi<sub>2</sub>. There have been some discussions and arguments<sup>4,5</sup> on a crossover from a surface-scattering-dominated regime to a thick-film regime by adding an impurity-induced contribution to scattering rates. However, so far, it has not been quite clear what the explicit connection between the quasiclassical and quantum approaches is and whether or not the quasiclassical theory can be reformed to be applicable to the ultrathin metallic films where the surface scattering dominates. Obviously, a rigorous, transparent, and unified treatment on such an issue is greatly desirable.

In this paper, we will make such an effort and address the above fundamental questions. Starting from a model Hamiltonian including the scattering potentials due to impurities and both two surfaces, we apply the Green's function approach and the Kubo formula in real space to give a microscopic quantum-mechanism description of the in-plane conductivity  $\sigma$  in metallic films. The obtained result for  $\sigma$  is found to be very similar in form to the Fuchs-Sondheimer formula so as to provide a direct comparison between the quasiclassical and quantum approaches. It then follows that the quantum result of  $\sigma$  could be obtained from an improved quasiclassical approach by taking into account a quantized nature of partical states and by introducing a novel treatment of the surface via angle-dependent specularity parameters. Moreover, the present result is shown to be applicable over a rather wide range of film thickness, exhibiting a natural crossover from a classical thick-film regime  $(d > \lambda)$  to an ultrathin film  $(d \ll \lambda)$  where quantum size effects are manifestly important. In addition, the additive rule for the conductivity is briefly discussed.

Let us consider an ideal metallic film of thickness d with perfect surfaces perpendicular to the z axis and defined by the equations z = 0 and z = d. The Hamil-

tonian of a charge carrier confined in the film may be written as  $H_0 = \mathbf{p}^2/2m + V[\Theta(-z) + \Theta(z-d)]$ , where V is the confining potential height and  $\Theta(z)$  is the unit step function. Eigenenergies of  $H_0$  are given by  $E_{\mathbf{k},n} =$  $(\mathbf{k}_{\parallel}^2+q_n^2)/(2m)$ , where the in-plane momentum  $\mathbf{k}_{\parallel}$  is continuous and the out-of-plane momentum  $q_n = n\pi/d$  is discrete with the subband index n taken to be 1,2,.... If assuming that the carrier wave functions are almost entirely confined inside the film, we can take V as infinite and then  $\Psi_n(z) = (2/d)^{1/2} \sin(q_n z)$ . The unperturbed one-particle Green's function is given by  $G_0(\mathbf{k}_{\parallel},n) =$  $\langle \mathbf{k}_{||}, n | \hat{G} | \mathbf{k}_{||}, n \rangle = 2m/(\bar{k}^2 - q_n^2) \text{ with } \bar{k} = (k_F^2 - k_{||}^2)^{1/2}$ (where, in consideration of transport properties at low temperatures, the electron's energy has been taken to be the Fermi energy  $\varepsilon = \varepsilon_F = k_F^2/2m$ ). The scattering Hamiltonian giving rise to resistivity is

$$H' = \sum_{\alpha} U \delta(\mathbf{r} - \mathbf{R}_{\alpha}) + \sum_{\ell=1,2} V f_{\ell}(\mathbf{r}_{\parallel}) \delta(z - z_{\ell}).$$
 (1)

Here the first term describes the scattering by impurities or defects within the film,<sup>9</sup> in which  $\mathbf{r} \equiv (\mathbf{r}_{\parallel}, z)$ with  $\mathbf{r}_{\parallel}$  the vector in the x-y plane and  $\mathbf{R}_{\alpha}$  is the position of an impurity or defect. The second term stands for the scattering due to rough surfaces, 6 in which  $z_{\ell}$ is the position the  $\ell$ th surface  $(z_1 = 0 \text{ and } z_2 = d)$ and  $f_{\ell}(\mathbf{r}_{\parallel})$  represents the random surface roughness with  $f_{\ell}(\mathbf{r}_{\parallel}) \ll d$ . H' is usually treated as a perturbated interaction and here assumed to be switched in two steps. We first switch the impurity scattering within the film. The imaginary part of the one-particle self-energy for the impurity-averaged functions is given by  $\varepsilon_F/(k_F\lambda)$  with  $\lambda$ the electron's mean free path due to impurity scattering. For a thin film system  $\lambda$  differs slightly from the bulk mean free path  $\lambda_0$  by  $\lambda/\lambda_0 = \kappa/(n_c + 1/2)$ , where  $n_c = \text{Int}[\kappa]$  is the integer part of  $\kappa = k_F d/\pi$ . Consequently, the impurity-averaged Green's function is obtained as  $\bar{G}(\mathbf{k}_{\parallel}, n) = 2m/(k^2 - q_n^2)$ , where k is a complex wave number or propagation constant, which is given by  $k = (\bar{k}^2 + ik_F/\lambda)^{1/2}$ . For ease of perturbated calculation of the surface scattering we transform  $\tilde{G}(\mathbf{k}_{\parallel},n)$ into  $\bar{G}(\mathbf{k}_{\parallel},z,z')$  [abbreviated to  $\bar{G}(z,z')$  hereafter] in the mixed Bloch-Wannier representation, yielding

$$\bar{G}(z,z') = \frac{m}{\bar{k}} \frac{\cos[k(d-\tilde{z}_{-})] - \cos[k(d-\tilde{z}_{+})]}{\sin(kd)}, \quad (2)$$

with  $\tilde{z}_{\pm}=|z\pm z'|$ . Here, following Camblong and Levy, we have made the approximation  $k\simeq \bar{k}+ik_F/(2\bar{k}\lambda)$ , which is reasonable for most of the effective transport range with  $k_F\lambda\gg 1$ . In the limit of an infinite V,  $\Psi_n(z_\ell)$  at the surfaces vanishes, but  $V^{1/2}\Psi_n(z_\ell)$  has a finite value. It is easy to vertify that  $V^{1/2}\Psi_n(0)=(-1)^{n-1}V^{1/2}\Psi_n(d)=q_n/(md)^{1/2}$  so that we have  $V\bar{G}(z_\ell,z_{\ell'})=\bar{k}\cot(kd)$  for  $\ell=\ell'$  and  $-\bar{k}/\sin(kd)$  for  $\ell\neq\ell'$ , and  $V^{1/2}\bar{G}(z,z_\ell)=V^{1/2}\bar{G}(z_\ell,z)=(2m)^{1/2}\sin[k(z-d)]/\sin(kd)$  for  $z_\ell=0$  and  $=-(2m)^{1/2}\sin(kz)/\sin(kd)$  for  $z_\ell=d$ .

The next step is to switch on adiabatically surface scattering and to calculate the Green's function  $G(z,z^\prime)$  by use of the Dyson equation

$$G(z,z')=ar{G}(z,z')+\int du dv ar{G}(z,u) \Sigma^s(u,v) G(v,z'), \ \ (3)$$

where  $\Sigma^s(u, v)$  stands for the irreducible self-energy due to the surface scattering. The calculation of the self-energy is standard. Its real part can be absorbed as a redefinition of the energy reference level and the imaginary part is given by

$$\Sigma^{s}(\mathbf{k}_{\parallel}, z, z') = -iVQ\delta(z - z') \sum_{\ell} \delta(z - z_{\ell}), \qquad (4)$$

$$Q = -\mathrm{Im} \int rac{d^2 \mathbf{q}_{\parallel}}{(2\pi)^2} \langle |f|^2 \rangle [VG(\mathbf{q}_{\parallel}, z_{\ell}, z_{\ell})], \quad .$$
 (5)

where  $\langle |f|^2 \rangle$  characterizes the surface roughness<sup>6</sup> and the angular brackets indicate an average over all surface profiles. In deriving Eq. (5) we have used a white noise surface profile for the uncorrelated atomically rough surface,  $^{4-6,8}$  i.e., we take  $\langle |f_\ell(\mathbf{k}_{\parallel} - \mathbf{q}_{\parallel})|^2 \rangle = \langle |f|^2 \rangle$ , to be a constant independent of the in-plane momentum. A self-consistent calculation for Q will be given hereafter. At this stage we simply replace  $G(\mathbf{q}_{\parallel}, z_{\ell}, z_{\ell})$  by  $G_0(\mathbf{q}_{\parallel}, z_{\ell}, z_{\ell})$  so that the integral over  $q_{\parallel}$  is easily done and an approximate result for Q is given by  $Q_0 = (\pi^2/2d^3)\langle |f|^2 \rangle S(n_c)$  with  $S(n_c) = \sum_{n=1}^{n_c} n^2 = n_c(n_c+1)(2n_c+1)/6$ . Substituting Eqs. (4) and (5) into Eq. (3), taking into account the expressions for  $V\bar{G}(z_{\ell}, z_{\ell'})$ ,  $V^{1/2}\bar{G}(z, z_{\ell})$ , and  $V^{1/2}\bar{G}(z_{\ell}, z)$ , which have been given above, and after a lengthy algebra, we finally arrive at

$$G(z,z') = \left(\frac{m}{i\bar{k}}\right) \frac{\Gamma(z,z')}{1 - RT(2d)},\tag{6}$$

with

$$R = \left(\frac{1 - \bar{k}Q_0}{1 + \bar{k}Q_0}\right)^2 \tag{7}$$

and  $\Gamma(z,z')=T(\tilde{z}_-)+RT(2d-\tilde{z}_-)-R^{1/2}[T(\tilde{z}_+)+T(2d-\tilde{z}_+)],$  where  $T(z)=\exp[i\bar{k}z-k_Fz/(2\bar{k}\lambda)].$  Comparing Eq. (6) with Eq. (2), one finds that if the surface scattering is absent, i.e., Q=0 and R=1, Eq. (6) is identical to Eq. (2). It then follows that the effect of the surface scattering is entirely reflected by an effective reflection parameter R, which is momentum dependent. Equation (6) for G(z,z') is suitable for all z and z' except for  $z=z_\ell$  or  $z'=z_\ell$ . By use of a similar calculation one can obtain the result of  $VG(\mathbf{q}_\parallel,z_\ell,z_\ell)$ . Insertion into Eq. (5) gives the following self-consistent equation:

$$Q = \frac{\langle |f|^2 \rangle}{2d} \sum_{n=1}^{n_c} \frac{q_n^2}{1 + q_n Q}.$$
 (8)

Obviously,  $Q_0$  used above is the lowest approximation of Q. It is valid on the condition that  $k_FQ \ll 1$ , which corresponds to the small roughness under consideration.

The two-point conductivity<sup>9</sup> can be calculated from the Kubo formula<sup>11</sup> in real space

$$\sigma(z,z') = rac{e^2}{2m^2} \int_{m{k}_{\parallel} < m{k}_F} rac{d^2 \mathbf{k}_{\parallel}}{(2\pi)^3} k_{\parallel}^2 [A(\mathbf{k}_{\parallel},z,z')]^2, \qquad (9)$$

where  $A(\mathbf{k}_{\parallel},z,z')=i[G(z,z')-G^*(z,z')]$  is the spectral function of the one-particle Green's function and the average conductivity is given by  $\sigma=\langle\sigma(z,z')\rangle_d=d^{-1}\int_0^d dz\int_0^d dz'\sigma(z,z')$ . It can be shown that terms such as GG and  $G^*G^*$  have no contribution to the conductivity and so  $A^2$  in the integrand can be replaced by  $2GG^*$ . Changing further the integration variable  $k_{\parallel} \to \bar{k}$  and introducing  $\tilde{E}_d=\exp(-k_Fd/\bar{k}\lambda)$ , we obtain

$$\sigma = \frac{e^2}{4\pi^2} \int_0^{k_F} d\bar{k} \left( \frac{k_F^2 - \bar{k}^2}{\bar{k}} \right) \frac{\langle |\Gamma(z, z')|^2 \rangle_d}{1 - 2R\tilde{E}_d \cos(2\bar{k}d) + R^2 \tilde{E}_d^2}.$$
(10)

This equation for  $\sigma$  has several interesting features.  $\Gamma$ given above contains four terms, so  $|\Gamma|^2 = \Gamma \Gamma^*$  in the integrand may be divided into two parts: the sum of the modulus square of the individual term and the sum of all other crossed terms. We have found that the former contribution to the integral is much greater than the latter one. As a result, in the present calculation for  $|\Gamma|^2$ only the modulus squared terms need to be kept. Another crucial feature of Eq. (10) is that the integrand is an oscillatory function of period  $\pi/d$  and has minima at  $\bar{k} = q_n$ , analogous to the quantization condition of the wave vectors. It is convenient to divide the integration range into  $n_c$  small regions, respectively, between  $q_n - \pi/2d$  and  $q_n + \pi/2d$  so that the integral over  $\bar{k}$  in Eq. (10) can be replaced by  $\sum_{n=1}^{n_c} \int_{q_n-\pi/2d}^{q_n+\pi/2d} d\bar{k}$ . Since all the function in the integrand, except  $\cos(2\bar{k}d)$ , are slowly varying within each of the small integration regions, the argument  $\bar{k}$  of these functions can be approximately replaced by  $q_n$ . The remainder integration over  $\bar{k}$  is equal to  $[1-R^2\tilde{E}_d^2]^{-1}$ . Finally, we obtain

$$\frac{\sigma}{\sigma_0} = 1 - \frac{\lambda}{2\pi k_F N_e d^2} \sum_{n=1}^{n_c} q_n (k_F^2 - q_n^2) \frac{(1-R)(1-E_d)}{1-RE_d},$$

where  $E_d = \exp(-k_F d/q_n \lambda)$ ,  $\sigma_0 = N_e e^2 \lambda/k_F$  is the conductivity in the absence of surface scattering, and  $N_e = (2\pi d)^{-1} \sum_{n=1}^{n_c} (k_F^2 - q_n^2)$  is the carrier density. Equation (11) is the major result of this work, which

Equation (11) is the major result of this work, which provides a unified formula for the conductivity in metallic films. It is interesting to notice that this equation derived from a quantum method looks very much like the Fuchs-Sondheimer result,<sup>2</sup> since both of them seem to have exactly the same form. However, there are several important differences between them. In the Fuchs-Sondheimer treatment the surface scattering is characterized by introducing phenomenologically a reflection coefficient R, which is assumed to be a constant independent of the momentum of electrons. In the present work R is derived microscopically and shown to be momentum dependent, as given in Eq. (7). Another important difference is that the present out-of-plane momentums are discrete  $(q_n = n\pi/d)$ , while those in the quasiclassical theory are continuous, the former exhibiting a quantized nature of

particle states in a thin film. From the similarity in form between them, a suggestion follows that Eq. (11) could also be obtained from a quasiclassical approach in which the out-of-plane momentum is considered to be quantized and R is assumed to have the form of Eq. (7). The quasiclassical approach, which overcomes the shortcoming of omitting the quantum effects, is applicable to the ultrathin films of  $d \ll \lambda$ .

In the thick-film limit the quantum effect can be neglected and Eq. (11) should reduce to the quasiclassical result. To show this point let  $u=q_n/k_F$  give the cosine of the angle of incidence of the electron on the surface. For large d, or equivalently for  $k_Fd\gg 1$ , the discreteness in the eigenvalues is washed out so that u becomes a continuous variable and the summation over n is approximated by an integral over u between limits 0 and 1 ( $k_F \simeq n_c d/\pi$  for  $n_c \gg 1$ ). It then follows that  $N_e = k_F^3/(3\pi^2)$ , which is a well-known formula for the bulk electron density, and Eq. (11) becomes

$$\sigma = \sigma_0 \left( 1 - \frac{3\lambda}{2d} \int_0^1 u(1 - u^2) \frac{(1 - R)(1 - E_d)}{1 - RE_d} \right), \quad (12)$$

with  $E_d = \exp(-d/\lambda u)$ . Equation (12) is found to be the same as the Fuchs-Sondheimer result,<sup>2,10</sup> except that the present R has an angle dependence which is given by  $R = [(1 - uk_F Q)/(1 + uk_F Q)]^2$ .

We wish to emphasize that Eq. (11) is a general quantum formula for  $\sigma$  in metallic films because it is derived from a standard Green's function approach and the Kubo formula. It implies that Eq. (11) should also correctly describe the quantum effect of the transport in thin films discussed previously. To clarify this point, we consider the surface-scattering-dominated region of  $d \ll \lambda$  and rewrite Eq. (11) as

$$\sigma = \frac{e^2}{2\pi} \sum_{n=1}^{n_c} \frac{k_F^2 - q_n^2}{q_n} \left( \frac{1}{\alpha_n} - \frac{(1-R)(1-E_d)}{\alpha_n^2 (1-RE_d)} \right), \quad (13)$$

where  $\alpha_n = k_F d/q_n \lambda$  can be regarded as small parameters for any n as long as the film is sufficiently thin such that  $\alpha_1 < 1$ . In this case  $E_d$  is approximated by  $1 - \alpha_n + (1/2)\alpha_n^2$  and so the factor in large parentheses in Eq. (13) is approximately equal to  $(1/2)[(1+R)/(1-R+\alpha R)]$ . On the other hand, for small surface roughness under consideration, Q is also a small parameter so that  $R \simeq 1 - 4q_nQ_0$  to the lowest order in the surface roughness. Then, this factor is further approximated by  $[4q_nQ_0 + \alpha_n]^{-1}$ . As a result, Eq. (13) is reduced to

$$\sigma \simeq \frac{e^2}{2\pi k_F d} \sum_{n=1}^{n_c} \frac{k_F^2 - q_n^2}{\lambda^{-1} + 4q_n^2 Q_0/(k_F d)}.$$
 (14)

We now compare Eq. (14) with the result for  $\sigma$  obtained by FC (Ref. 6) and by TA.<sup>5</sup> First, consider the surfaceinduced resistivity of ultrathin metallic films by neglecting the impurity scattering, i.e., taking  $\lambda^{-1} = 0$  in the denominator in Eq. (14). In this case a substitution of  $Q_0$  into Eq. (14) gives

$$\sigma \simeq \frac{e^2 d^5}{4\pi^5 \langle |f|^2 \rangle S(n_c)} \sum_{n=1}^{n_c} \frac{k_F^2 - q_n^2}{n^2}.$$
 (15)

Taking into account that  $\langle |f|^2 \rangle = \Delta^2 \xi^2 F(0), n^2 = v^2,$  and  $k_F^2 - q_n^2 = k_v^2$ , where the parameters on the right-hand side of each equality are given by FC, 6 one finds that Eq. (15) is identical to Eq. (13) in Ref. 6, except for an additional prefactor 1/2. The present reduction in  $\sigma$  by a factor of 2 stems from the fact that we consider two rough surfaces as shown in Eq. (1), while FC assumed only one nonideal surface to exist. Since Eq. (15) is the approximate result for  $\sigma$  to the lowest order in the surface roughness, the scattering effect of two rough surfaces is simply additive.

Next, we consider the resistivity due to both impurity and surface scattering. It was suggested<sup>5</sup> that in the presence of both impurity and roughness the total scattering rate for an electron in the nth subband is given by a sum of the impurity and surface scattering rates, the latter depending on the subband index n. For simplicity of comparison, their result for  $\sigma$ , given by Eq. (4.13) of Ref. 5, is rewritten as

$$\sigma \simeq \frac{e^2}{2\pi k_F d} \times \sum_{n=1}^{n_c} \frac{k_F^2 - q_n^2}{(2n_c + 1)/(2\kappa\lambda_0) + \pi n^2 (\delta d/d)^2 S(n_c)/(n_c^3 d)}.$$
(16)

It is straightforward to find the equivalence between Eqs. (14) and (16). First, their prefactors and both the numerators in the summations are exactly same. Second, the first term in both denominators are also same, since  $\lambda^{-1} = (n_c + 1/2)/\kappa \lambda_0$  is just the size dependence of  $\lambda$  in a ultrathin film, which has been mentioned above. The remainder is a comparison between the second terms in both denominators, which indicate the inverse mean free path for the nth subband due to the surface roughness. Their equivalence is shown to need two equalities:

 $n_c=k_Fd/\pi$  and  $(\delta d)^2=k_F^2\langle|f|^2\rangle$ . The former is a reasonable approximation suitable for metellic films where  $n_c\gg 1$  and the latter can be regarded as a corresponence of different parameters used to characterize the surface roughness. With the aid of the two equalities these two terms in Eqs. (14) and (16) are found to coincide with each other. The only difference is that the present surface-induced mean-free-path is one half of that given by TA,<sup>5</sup> where a rough surface alone is considered.

Equation (14) shows that for each subband the scattering rate due to impurity and surface is additive and the total conductivity is given by a sum of conductivities of all the subbands. This result is obtained in the lowest approximation of the surface roughness. In general, owing to the effect of intersubband scattering by the surface roughness, the total conductivity cannot be written as a simple form such as Eq. (14), even though the total selfenergy is given by a sum of the two independent parts due to the impurities and surfaces. This is a character of a thin-film system with quantized out-of-plane wave vectors, which is quite different from bulk systems including multilayers where the plane wave functions can be used. It is believed that Eq. (11) derived here includes this effect and can describe correctly the crossover behavior from the surface-dominated regime to the thick-film limit where the impurity scattering dominates. The detailed discussion will be presented elsewhere.

In summary, we have developed a unified transport theory of metallic films. A general formula for the conductivity has been derived more rigorously in the presence of both impurity scattering and surface roughness. It reveals the connection between the quasiclassical and quantum theories and gives a natural crossover from the classical to quantum result.

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