Quantum Transport and Surface Scattering

Zlatko Tešanović and Marko V. Jarić[a]
Lyman Laboratory of Physics, Harvard University, Cambridge, Massachusetts 02138
and
Sadamichi Maekawa[b]
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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The effects of surface scattering on quantum transport in thin films and wires are considered. A nontrivial channel-mixing problem arising from random boundary conditions is solved exactly by a new method that we present here. We find that the full quantum-mechanical treatment of very pure systems leads to results qualitatively different from those of currently used quasiclassical theory. Our theory explains some recent experimental observations.

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Recent advances in microfabrication of metallic and semiconductor nanostructures have led to samples whose miniature dimensions and high purity allow for a direct observation of quantum size effects.[1-3] In such samples the mean free path due to ordinary impurity scattering can be orders of magnitude longer than the thickness or radius of a sample. Under these conditions any realistic discussion of quantum transport in thin films and wires has to take into account the effects of surface scattering. It is therefore of great practical interest to understand in detail the limitations on quantum transport arising from the scattering off rough boundaries.

At present such understanding is not available. In most of the experimental work the size effects in the electrical conductivity are included via *quasiclassical* formulas of Fuchs, Englman and Sondheimer, Ziman, and Soffer[4] derived by consideration of the Boltzmann transport in the presence of a diffusely reflecting boundary. While these formulas seem to work well for thicker, relatively dirty systems[5] they are completely inadequate for very thin, high-purity samples. This becomes apparent if one considers the conductivity σ of a film of thickness d with diffusely reflecting surface. In the limit when the impurity mean free path l goes to infinity $\sigma \propto d \ln(l/d)$ (Ref. 4) diverges, an implication that in the absence of a bulk relaxation mechanism the scattering off the random surface induces *no dissipation* of electrical current. This nonphysical result is a direct consequence of the complete omission of quantum size effects from the quasiclassical theory.

In this Letter we present the fully quantum theory of transport in films or wires with rough surfaces. We show how to relate the variations in the surface profile to a set of pseudopotentials acting on the quantum states of a system with the same average thickness or radius but with a smooth surface. The method for evaluation of these pseudopotentials is quite general and can be applied to simple confining potentials (impenetrable box or harmonic oscillator) as well as to some effective confining potential resulting from a more microscopic calculation.

We define boundary roughness as variation in the parallel space ($x_\parallel$) of a single length scale a of some confining potential $U_a(x_\perp)$ in the perpendicular space ($x_\perp$). The full Hamiltonian for this problem can be written as $H = H_0(x_\parallel) + H_{a(x_\parallel)}(x_\perp)$, where H_0 describes extended states in the parallel space, $H_{a(x_\parallel)} \equiv -(\hbar^2/2M)\nabla_\perp^2 + U_{a(x_\parallel)}(x_\perp)$, and subscript $a(x_\parallel)$ denotes the variable length scale of the confining potential. Here we are interested in deviations of $a(x_\parallel)$ from some average a. Assuming that these deviations are small, we can formulate perturbative treatment of H.

Consider the operator $\exp S(x_\parallel) \equiv \exp\lambda(x_\parallel)\exp S_0(x_\parallel)$, where $S_0(x_\parallel) = \frac{1}{2}\lambda(x_\parallel)[x_\perp(\partial/\partial x_\perp) + (\partial/\partial x_\perp)x_\perp]$ and $\lambda = \ln[a/a(x_\parallel)]$. While the *unitary* operator $\exp S_0$ acting on the eigenstates $|\Psi_a\rangle$ of H_a generates the eigenstates $|\Psi_{a(x_\parallel)}\rangle$ of $H_{a(x_\parallel)}$, the *nonunitary* operator $\exp S$ acting on H_a generates $H_{a(x_\parallel)}$, that is

$$H_{a(x_\parallel)} = \exp[S(x_\parallel)]H_a\exp[S^\dagger(x_\parallel)] = \exp[2\lambda(x_\parallel)]\exp[S_0(x_\parallel)]H_a\exp[-S_0(x_\parallel)]. \quad (1)$$

Equation (1) now enables systematic evaluation of the effective Hamiltonian that will serve as a basis for the perturbation expansion. To the leading order in λ, which is the small parameter of the theory, this effective Hamiltonian is characterized by the set of channel-mixing pseudopotentials $2\lambda\langle\Psi_a'|H_a|\Psi_a''\rangle + \langle\Psi_a'|[S_0,H_a]|\Psi_a''\rangle$.

Up to this point the discussion was completely general. We now focus on the specific case of a film with a rough surface parallel to the x-y plane: then $x_\parallel \equiv (x,y)$ and $x_\perp \equiv z$.[6] Using the leading order expansion of (1), and assuming that the particles move freely in the x-y plane, we can write the effective Hamiltonian for the rough surface problem:

$$H_{\text{eff}} = \sum_{\mathbf{k},n}(\xi_{\mathbf{k}} + E_n)c_{\mathbf{k}n}^\dagger c_{\mathbf{k}n} + \sum_{\mathbf{k},\mathbf{q}}\sum_{n,m}\lambda(\mathbf{q})V_{nm}c_{\mathbf{k}+\mathbf{q}n}^\dagger c_{\mathbf{k}m}. \quad (2)$$

Here $\mathbf{k}=(k_x,k_y)$ and $\lambda(\mathbf{q})$ is a Fourier transform of $\lambda(x,y)$. The matrix V_{nm} is defined as

$$V_{nm}=2E_n\delta_{nm}+\tfrac{1}{2}(E_m-E_n)[z\,\partial/\partial z+(\partial/\partial z)z]_{nm}, \quad (3)$$

where E_n are the eigenvalues of $H_a(z)$.

The effective Hamiltonian (2) can be treated by a familiar perturbation theory.[7] The channel-mixing pseudopotential $\lambda(\mathbf{q})V_{nm}$ is directly related to variations in the surface profile and the explicit form of V_{nm} depends on a particular choice for the confining potential. It is clear that the self-energy matrix W_{nm} and the full Green's function G_{nm} cannot be calculated for any V_{nm}. To evaluate G_{nm} for an arbitrary confining potential $U(z)$ is is necessary to invert an $n_c\times n_c$ matrix, n_c being the number of subbands in the $\mathbf{k}$ space located at the Fermi level. As n_c is a large number for most cases of interest, it appears that finding G_{nm} and carrying out the summation of the perturbation series for conductivity is a highly impractical task.

However, in the special case of a particle-in-a-box potential, G_{nm} can be found explicitly for any n_c. This is possible because in this case V_{nm} is a separable matrix, i.e., $V_{nm}=f_nf_mV$, where $f_n=n(-)^n$ and $V=\hbar^2\pi^2/Md^2$. The separability of V_{nm} enables us to treat the matrix character of a perturbation expansion *exactly*. Although $U(z)$ is nonanalytic in this case, V_{nm}, as defined in (3), is perfectly well behaved. This makes it possible to discuss this potential with use of the effective Hamiltonian (2), on equal footing with other confining potentials of which indeed it is a special case.[8] Defining $w(x,y)=\lambda(x,y)d$, where d is the average thickness, we find the self-energy $W_{nm}(\mathbf{k},\omega)=f_nf_mW(\mathbf{k},\omega)$ and the Green's function

$$W(\mathbf{k},\omega)=\frac{\hbar^4\pi^4}{M^2d^6}\sum_{\mathbf{q}}\langle w(\mathbf{q})w(-\mathbf{q})\rangle\frac{g_0(\mathbf{k}-\mathbf{q},\omega)}{1-W(\mathbf{k}-\mathbf{q},\omega)g_0(\mathbf{k}-\mathbf{q},\omega)}, \quad (4)$$

$$G_{nm}=G_{0n}\delta_{nm}+G_{0n}[f_nf_mW/(1-Wg_0)]G_{0m}, \quad (5)$$

where $g_0\equiv\sum_nf_n^2G_{0n}$ and argument $(\mathbf{k},\omega)$ is understood.

We are now in the position to calculate the conductivity of a film as limited only by surface scattering. This can be done for an arbitrary surface profile as long as the second moment is well defined, since the separability enables exact summation of the ladder diagrams. In particular, the effects of correlations in surface irregularities can be included, leading to reduced scattering rate. This offers a possibility of a "first-principle" calculation of the conductivity when there is independent information regarding $\langle w(\mathbf{q})w(-\mathbf{q})\rangle$, a quantity that is directly measurable by scanning tunneling microscopy, for example. For simplicity, we consider here the "white noise" surface profile describing an uncorrelated, atomically rough surface. This implies $\langle w(\mathbf{q})w(-\mathbf{q})\rangle=a^2H^2$, where $a\sim k_F^{-1}$ and H^2 measures the rms fluctuations in film thickness. The conductivity is calculated from a Kubo formula[7] allowing for the matrix character of Green's functions. The leading term in H^2 comes from a single loop, i.e., $\sigma'\propto\mathrm{Tr}(G^RT^A)$. For ordinary dirty metals this term gives the classical, Boltzmann result. In our case, for $n_c\cong k_Fd/\pi\gg1$,

$$\sigma'=\frac{e^2}{\hbar}6\pi s(n_c)\frac{n_c^3}{k_F^2H^2}\left[1+O\left(\frac{k_F^4H^4}{n_c^2}\right)+\cdots\right], \quad (6)$$

where $s(n_c)=\sum_{n=1}^{n_c}(1/n^2)\simeq\pi^2/6$.

$\rho'=\sigma'^{-1}$ represents the residual resistivity due only to the boundary scattering. It has *no classical analog*. As $\hbar\to0$, ρ' vanishes, unlike its counterpart for impurity scattering. The zero resistivity arises since, classically, it is possible to prepare a beam of electrons moving parallel to a rough surface and traveling ballistically across the sample. The quantum-mechanical zero-point motion excludes momentum states confined to the x-y plane and results in finite resistivity. That this is the case can be seen from the effective mean free path deduced from σ', $l'=6\pi(n_c)(n_c^2/k_F^2H^2)d$. This indicates that conductivity is dominated by the longest mean free path, $l_{\max}=6\pi(n_c^2/k_F^2H^2)d$, corresponding to the $n=1$ subband and accounts for rather strong d^3 dependence of σ'. Note that l' is not of order d and can be much longer.

Except for the highest-purity samples it will be necessary to consider the interference between impurity and surface scattering. In this context, of particular interest are the size effects describing the increase in resistivity of a film as d becomes shorter than the impurity mean free path l. Using our theory, we can calculate these effect including impurities as an additional source of scattering in Eq. (2). Defining σ_∞ as conductivity of a bulk system, we obtain

$$\frac{\sigma'}{\sigma_\infty}=\frac{1}{n_c}\sum_{n=1}^{n_c}\left[1+\frac{l}{l_{\max}}n^2\right]^{-1}. \quad (7)$$

The expression (7) describes the crossover from the impurity-scattering-dominated region ($d/l\gg1$) to the thin-film ($d/l\ll1$), where the surface scattering dominates. In Fig. 1 we test (7) against recent experimental results of Hensel *et al.*,[1] where the resistivity of epitaxially grown, single-crystal $CoSi_2$ films has been measured as a function of thickness in a wide region, $60\le d\le1100$ Å. These are some of the best-quality samples, with long mean free path, $l\simeq1000$ Å. The full line is the best fit to the data of Eq. (7), obtained for $H=2.06$ Å, a value that appears to be quite reasonable from what is known about these films.[9] For $\kappa=d/l>0.2$, it basically coincides with the $p=0.9$ quasiclassical curve. However, for $\kappa<0.2$ there is a qualitative change and

the resistivity increases much faster than predicted by quasiclassical formulas reflecting the trend obvious in the experimental results. It appears that Eq. (7) provides a better overall fit than either of the dashed lines. As high-quality single-crystal films with smaller κ become available the qualitative difference between (7) and quasiclassical expressions will become even more apparent.

Further experimental evidence that Eq. (7) is relevant for thin films comes from the work of Orr *et al.*,[10] who studied very thin Sn films grown by deposition on a cold substrate. A crossover in resistivity from standard $\rho \propto 1/d$ to $\rho \propto 1/d^{\nu}$, where $\nu \simeq 2$, has been observed for very small d. We can explain this result by considering the crossover predicted by Eq. (7) in the limit of very thin films, where surface scattering plays a decisive role. Theoretical models of film growth imply $H^2 \propto d$ for randomly grown film and $H^2 \propto d^{2/3}$ for ballistic deposition. From Eq. (6) we predict that $\nu = 2$ and $\nu = \frac{7}{3}$, respectively, for these two models of surface profile, both within the fitting error of experimental ν.

From Hamiltonian (2) it seems clear that surface roughness may induce weak localization effects in thin films even in the absence of any bulk relaxation mechanism. In ordinary dirty systems the origin of localization lies in enhanced backscattering.[7,11] We can study these effects by considering the sum of "maximally crossed" diagrams.[7] Again, these diagrams cannot be summed for an arbitrary confining potential, but for particle-in-a-box the *Ansatz* $D_{nmrs}(\mathbf{q},\omega) = f_n f_m f_r f_s D(\mathbf{q},\omega)$ enables us to find Cooperon propagator in a closed form:

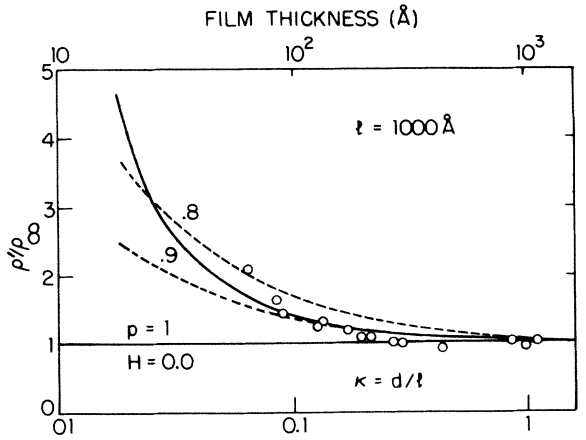

FIG. 1. Resistivity vs thickness for fourteen thin-film samples of $CoSi_2$ from the experiment of Hensel *et al.* (circles). Solid line represents (ρ'/ρ_∞) as found from Eq. (7) with $H = 2.06$ Å. Two dashed lines resulting from quasiclassical theory correspond to two values of specularity parameter p.

$$D(\mathbf{q},\omega) = V^2\left[1 - V^2 \sum_{n,m,r,s,\mathbf{k}} f_n f_m f_r f_s G^R_{nm}(\mathbf{k}+\mathbf{q},\omega) G^A_{rs}(-\mathbf{k},\omega)\right]^{-1}. \tag{8}$$

With the help of Eqs. (4) and (5) one can prove that $D(\mathbf{q},\omega)$ possesses a diffusion pole as $q \to 0$, $\omega \to 0$. Expansion around this pole and summation over channel indices produce a scale-dependent correction to σ':

$$\sigma'' = -\frac{e^2}{\hbar \pi^2} \sum_{n,m} A_{nm} \ln \frac{L}{l_{nm}} \left(\sum_{n,m} A_{nm}\right)^{-1}, \tag{9}$$

where $l_{nm} = l_{\max}/nm$ and

$$A_{nm} = \frac{2\pi\tau}{\hbar} \sum_{n,m} \frac{n^2+m^2}{(E_n - E_m)^2 + (\hbar^2/4\tau^2)(n^2+m^2)^2} \tag{10}$$

with $\tau \equiv l_{\max}/v_F$. Therefore, we arrive at the result that, even in the absence of impurities, all states in an infinite film are localized at $T = 0$, for arbitrary large thickness and for arbitrary small but finite surface roughness.[12] If the upper cutoff at nonzero temperature is some dephasing length $L_\phi \propto T^{-p/2}$, Eq. (9) gives $\sigma'' = (e^2/2\pi^2\hbar) \times p\alpha_T \ln T$ with $\alpha_T = 1$, same as for ordinary localization.

The fact that surface roughness leads to $\alpha_T = 1$ can be used to explain the results of Chaudhari, Habermeier, and Maekawa.[2] They have observed that in a single-crystal Au films, α_T is not universal and varies with the sheet resistance of a sample, decreasing from 5.86 for $R = 8$ Ω to 1.6 at $R = 32$ Ω. These are extremely clean films, with impurity mean free paths in excess of 10^4 Å, and only sources of scattering are thought to be the surface roughness and dislocations developed during the manufacturing process.[13] The dislocations are preferentially in the z direction and will not induce interband scattering. This would imply $\alpha_T \simeq n_c$. The interference of these two processes results in α_T in between 1 and n_c and reproduces the qualitative trend found experimentally if the increase in resistance is correlated with rougher surfaces and thinner films.[14] The quantitative fit can actually be made very precise but we will not discuss it here for a lack of space. It is important to emphasize that this mechanism offers *an alternative* to explanation of nonuniversality by the Kondo effect.[15] The Kondo effect is not important in thin films ($d/l \ll 1$). If one assumes that the impurity mean free path l is proportional to $\ln T$ because of the Kondo effect, it is obvious from (7) that this $\ln T$ dependence will not be reflected in the resistivity.

In summary, we have developed the theory of transport in small structures with rough surfaces, taking into account a quantized nature of particle states and treating the channel mixing exactly within the perturbation expansion. Our results represent qualitative improvement over currently used quasiclassical theory. We have demonstrated that our theory accounts very well for several recent experimental observations.

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[a]Present address: Center for Theoretical Physics, Texas A&M University, College Station, TX 77843.

[b]Permanent address: The Research Institute for Iron, Steel, and Other Metals, Tohoku University, Sendai 980, Japan.

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