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LETTER TO THE EDITOR

Surface-induced resistivity of gold films on mica: comparison between the classical and the quantum theory

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Abstract. We report an extension of the theory of Sheng, Xing and Wang (SXW) (Sheng L, Xing D Y and Wang Z D 1995 *Phys. Rev. B* **51** 7325), which permits the calculation of size effects from the statistical properties that characterize the surface on a microscopic scale, for samples in which the average height–height autocorrelation function (ACF) is described either by a Gaussian or by an exponential. We also report measurements of the topography of a gold film deposited on a mica substrate using a scanning tunnelling microscope (STM) on a gold sample 70 nm thick deposited under ultrahigh vacuum on a mica substrate preheated to 300 °C. From the STM images we compute the average ACF which characterizes the surface of the film on the scale of 10 nm × 10 nm, and determine by least-squares fitting the r.m.s. amplitude δ and the lateral correlation length ξ corresponding to a Gaussian and to an exponential that best represent the ACF data. Using the modified SXW (mSXW) theory and a Gaussian and an exponential representation of the ACF data, we calculate the quantum reflectivity R characterizing the interaction between the electrons and the surface, and the decrease in conductivity $\Delta\sigma$ attributable to electron–surface scattering, for mean free paths $2.5 \text{ nm} \leq \ell \leq 1000 \text{ nm}$. We compare the predictions of the classical Fuchs–Sondheimer (FS) model for the average quantum reflectivity $R = \langle R \rangle$, calculated with the mSXW model, with the predictions of the quantum theory, using both the Gaussian and the exponential representation of the ACF. We find that $\Delta\sigma$ predicted by FS theory for $R = \langle R \rangle$ exceeds that predicted by the quantum mSXW theory, by an amount that increases with increasing ℓ . This discrepancy can be traced to the angular dependence of the quantum reflectivity $R[\cos(\theta)]$. We also find that the decrease in conductivity $\Delta\sigma$ predicted by mSXW theory for a Gaussian representation of the data is larger than that predicted for an exponential representation of the *same ACF data*. We attribute this to the fact that the reflectivity R is determined by the Fourier transform of the ACF, and the Gaussian and the exponential that best represent the ACF data exhibit Fourier transforms that are similar in the regions where $k\xi \sim 1$, but are different in the regions where $k\xi < 1$ and $k\xi > 1$ (k : wave vector).

One of the fundamental problems in solid-state physics, which has attracted the attention of researchers for over fifty years, relates to the effect of electron–surface scattering on the transport properties of thin metallic and semiconducting films—what is known as ‘size effects’. The theoretical work concerning size effects focused for many decades on the use of classical models based on solutions of the Boltzmann transport equation, where the effect of the rough

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surface is incorporated into the boundary conditions that must be satisfied by the electron distribution function, via a specularity parameter R that represents the fraction of electrons, $0 \leq R \leq 1$, that are specularly reflected upon colliding with the rough surface [1].

It is well known that this approach is inadequate for very thin, high-purity samples, for which the film thickness t is much smaller than the impurity mean free path ℓ . In the limit where ℓ goes to infinity, the conductivity diverges as $\ln(\ell/t)$, an unphysical result that arises from the omission of quantum effects in the classical theory. To overcome these shortcomings, a number of transport theories applicable to several special cases have been published which provide a quantum mechanical description of transport [2–4]. The quantum transport theories succeeded in describing the experimental data on ultrathin films of CoSi_2 [5]. These theories were recently generalized by Sheng, Xing and Wang (SXW), who proposed a unified formulation that permits the calculation of the resistivity arising from electron–surface scattering in continuous films of arbitrary thickness [6]. It has been shown that:

- (a) in the thick-rough-film limit where quantum effects can be neglected and bulk scattering dominates over surface scattering, the SXW theory reproduces the classical result of Fuchs and Sondheimer (FS) [1, 6];
- (b) in the limit where surface and bulk scattering are comparable, the SXW theory reproduces the result of Trivedi and Ashcroft [3, 6];
- (c) in the limit of ultrathin films where surface scattering is expected to dominate over bulk scattering, the SXW theory reproduces the result of Fishman and Calecki [4, 6].

In their treatment, SXW modelled the surface roughness by a white-noise surface profile, assuming that the Fourier transform of the height–height autocorrelation function (ACF) that on average characterizes the surface is a constant independent of the in-plane momentum [6]. The SXW theory can be extended beyond the white-noise approximation to allow the calculation of the resistivity arising from electron–surface scattering *from first principles, from the information contained in the surface roughness profile*. A formalism that permits the calculation of the macroscopically observable size effects from the statistical properties that characterize the surface on a microscopic scale has been one of the goals of theoretical research for decades; consequently such an extension is highly desirable. In this letter we present an extension of SXW theory to samples in which the average ACF is represented either by a Gaussian or an exponential. We also present, for the first time, a comparison between the classical FS model and the quantum mSXW model for the range of mean free path $2.5 \text{ nm} \leq \ell \leq 1000 \text{ nm}$, using both a Gaussian and an exponential representation of the ACF. The ACF data were obtained from the surface topography of a gold film 70 nm thick deposited under UHV on a mica substrate preheated to 300 °C, measured with a scanning tunnelling microscope (STM).

To summarize results already published, the SXW formalism shows that the many-body quantum theory leads to a reformulation of the FS model that includes the effects of surface scattering via a reflectivity parameter R that can be calculated from equation (7) in reference [6]:

$$R[k_{\parallel}] = [(1 - k_z Q(k_{\parallel})) / (1 + k_z Q(k_{\parallel}))]^2$$

where $Q(k_{\parallel})$ represents the dissipative part of the self-energy of the electron gas due to electron–surface scattering and $k_z^2 = k_F^2 - k_{\parallel}^2$, where k_F stands for the Fermi momentum and $k_{\parallel} = (k_x, k_y)$ represents the in-plane momentum. The ratio of the film conductivity σ to the bulk conductivity σ_0 may be computed in terms of the reflectivity R :

$$1 - \frac{\sigma}{\sigma_0} = \frac{3}{2} \frac{\ell}{t} \frac{1}{X_0 N_c} \sum_{n=1}^{N_c} u_n (1 - u_n^2) \frac{(1 - R(u_n))(1 - E_d(u_n))}{1 - R(u_n)E_d(u_n)} \quad (1)$$

where t is the film thickness, ℓ the carrier mean free path, $u_n = q_n/k_F = \cos \theta_n = n\pi/(tk_F)$, $X_c = tk_F/\pi$, $N_c = \text{int}(X_c)$ where $\text{int}(M)$ stands for the integer part of M ,

$$X_0 = \frac{3}{2} \left[1 - \frac{1}{3} \left(\frac{N_c}{X_c} \right)^2 \left(1 + \frac{1}{N_c} \right) \left(1 + \frac{1}{2N_c} \right) \right]$$

and $E_d(u_n) = \exp[-t/(u_n\ell)]$, which corresponds to equation (11) of reference [6].

Following SXW, we consider, as a first approximation, the Green's function G_0 describing electrons bounded by two flat, parallel surfaces at $z = 0$ and $z = t$, for which $VG_0(k_z) = V_0G(k_{\parallel}) = k_z \cot(tk_z)$. To treat surface profiles as measured by a STM, we calculate the function $Q(k_{\parallel})$ for the case where the surface roughness *does not follow a white-noise law*. The constant $\langle |f|^2 \rangle$ appearing in equation (5) of reference [6] represents the Fourier transform of the ACF [4, 6], leading to

$$Q(k_{\parallel}) = -\text{Im} \int \frac{d^2q_{\parallel}}{(2\pi)^2} F(k_{\parallel} - q_{\parallel}) \bar{q} \cot(t\bar{q}) \quad (2)$$

where $\text{Im}(C)$ stands for the imaginary part of a complex number C , $\bar{q} = (q_z^2 + ik_F/\ell)^{1/2}$ is a complex wavenumber and $F(k_{\parallel})$ is the Fourier transform of the average height–height ACF $f(r_{\parallel})$ defined by

$$f(r_{\parallel}) = S^{-1} \int_S h(a_{\parallel}) h(a_{\parallel} + r_{\parallel}) d^2a_{\parallel} \quad (3)$$

where $r_{\parallel} = (x, y)$ stands for the in-plane coordinates and $h(a_{\parallel})$ represents the random height characterizing the surface roughness with respect to the average surface at $z = t$. Surface scattering at $z = 0$ has been ignored[†].

We compute the self-energy $Q(k_{\parallel})$ from equation (2), for the cases where the average ACF that characterizes the surface is described by a Gaussian

$$f(x, y) = \delta^2 \exp[-(x^2 + y^2)/\xi^2]$$

or by an exponential

$$f(x, y) = \delta^2 \exp[-\sqrt{x^2 + y^2}/\xi].$$

Since the corresponding Fourier transforms are real, the main contributions to $Q(k_{\parallel})$ arise from the poles of the function $\cot(t\bar{q})$, which can be evaluated using a Mittag–Leffler expansion of $\bar{q} \cot(t\bar{q})$ [7], leading to

$$Q(k_{\parallel}) = -\frac{\delta^2 \xi^2}{\pi t} \sum_{n=1}^{N_c} \left(\frac{n\pi}{t} \right)^2 \text{Im} \int_0^{\infty} \frac{g(k_{\parallel}, q_{\parallel}) q_{\parallel}}{k_F^2 - q_{\parallel}^2 - (n\pi/t)^2 + ik_F/\ell} dq_{\parallel}. \quad (4)$$

The integrand in equation (4) consists of a slowly varying real function $g(k_{\parallel}, q_{\parallel})$ (that depends on the choice of either a Gaussian or an exponential ACF) and the complex fraction $1/[k_F^2 - q_{\parallel}^2 - (n\pi/t)^2 + ik_F/\ell]$. The contributions to $Q(k_{\parallel})$ arise mainly from the values of q_{\parallel} that are in the neighbourhood of $q_{\parallel} = q_n$ such that the complex fraction is purely imaginary, e.g. $k_F^2 - q_n^2 - (n\pi/t)^2 = 0$; hence $q_n^2 = k_F^2 - (n\pi/t)^2$. The imaginary part of the integral in equation (4) can be approximated by evaluating the function $g(k_{\parallel}, q_{\parallel})$ at

[†] The mica substrate was freshly cleaved before loading the substrate and the gold in the UHV evaporation station. Because of the crystalline nature of the mica substrate verified via x-ray diffraction, the roughness of the mica consists of some rather infrequent cleavage steps. Consequently, surface scattering occurring at the surface of the mica, $z = 0$, can be safely ignored.

$q_{\parallel} = q_n$. In the case of a Gaussian ACF, these approximations, in the limit $\xi^2 k_F / (4\ell) \ll 1$, lead to

$$Q(k_{\parallel}) = \frac{\xi^2 \delta^2}{2t} \pi \exp \left[-\frac{\xi^2}{4} (k_{\parallel}^2 + k_F^2) \right] \sum_{n=1}^{N_c} \left(\frac{n\pi}{t} \right)^2 \times \exp \left[\left(\frac{n\pi}{t} \right)^2 \frac{\xi^2}{4} \right] I_0 \left(\frac{\xi^2}{2} k_{\parallel} \sqrt{k_F^2 - \left(\frac{n\pi}{t} \right)^2} \right) \quad (5)$$

where $I_0(x)$ stands for the modified Bessel function of order zero [8].

In the case of an exponential ACF we obtain, in the limit $\xi^2 k_F / \ell \ll 1$,

$$Q(k_{\parallel}) = \frac{2\xi^2 \delta^2}{t} \sum_{n=1}^{N_c} \left(\frac{n\pi}{t} \right)^2 \frac{E[r^2(k_{\parallel}, q_n)]}{[1 + \xi^2(k_{\parallel} - q_n)^2] \sqrt{1 + \xi^2(k_{\parallel} + q_n)^2}} \quad (6)$$

with

$$r^2(k_{\parallel}, q_n) = \frac{4\xi^2 k_{\parallel} q_n}{1 + \xi^2(k_{\parallel} + q_n)^2}$$

where $E(r^2)$ stands for the elliptic integral of the second kind [9].

Equations (1), (5) and (6) permit the calculation of the quantum reflectivity R and of the change in conductivity attributable to size effects, in terms of the r.m.s. amplitude δ and of the lateral correlation length ξ that describe the average ACF on a nanoscopic scale for either of the two models, Gaussian or exponential in a continuous film of thickness t .

We measured the surface topography of a gold film 70 nm thick deposited on a mica substrate preheated to 300 °C in UHV, on a scale of 20 nm \times 20 nm. Before imaging the gold sample, we verified that the freshly prepared W tip produced neat images of C atoms running on HOPG. From STM images we computed the average height–height ACF characterizing the gold film on a scale of 10 nm \times 10 nm, as the average of twenty ACFs calculated according to equation (3) from the roughness profiles recorded at *random* locations of the sample on a scale of 20 nm \times 20 nm using periodic boundary conditions, from twenty STM images containing 256 \times 256 pixels each. The resulting average ACF is displayed in figure 1. The peak at the origin of the average ACF was fitted using the Gaussian and the exponential models for the ACF, employing a least-squares fitting procedure. When the exponential was fitted to 6 \times 6, 8 \times 8 and 10 \times 10 pixels near the origin, the values obtained were [$\delta = 0.746$ nm, $\xi = 0.198$ nm], [$\delta = 0.687$ nm, $\xi = 0.231$ nm], [$\delta = 0.633$ nm, $\xi = 0.271$ nm]. When the Gaussian was fitted to 8 \times 8, 10 \times 10 and 12 \times 12 pixels near the origin, the values obtained were [$\delta = 0.494$ nm, $\xi = 0.401$ nm], [$\delta = 0.448$ nm, $\xi = 0.489$ nm], [$\delta = 0.422$ nm, $\xi = 0.549$ nm]. The values obtained for ξ and δ are consistent with the atomic resolution exhibited by the tip of the STM when running on HOPG prior to measurements on the gold sample; consequently the rounding off that could be expected on the images recorded with the STM due to the finite radius of curvature of the tip does not seem to play a significant role.

At this point, it seems appropriate to discuss the validity of the formalism used in mSXW theory, where the electrons are described by wave functions labelled by a wave vector k_z that is quantized as a consequence of confinement between two parallel potential barriers. However, the upper and lower surfaces of the metallic film are certainly *not* parallel to each other. Hence, the very existence of the subbands used in mSXW theory may seem questionable. This issue has been addressed in the literature. It has been found that a sufficient condition for the formation of subbands in a rough metallic film is that the upper and lower surfaces of the film

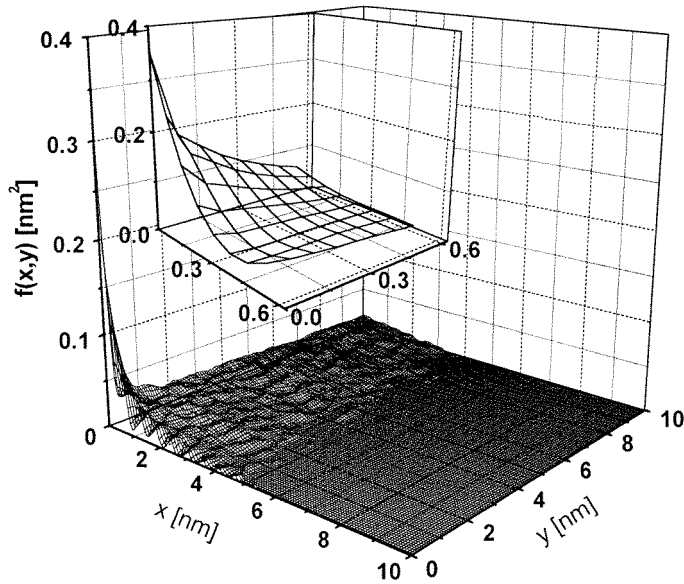


Figure 1. The average of twenty ACFs calculated from the surface roughness profiles recorded at random locations of the sample on a scale of $20 \text{ nm} \times 20 \text{ nm}$ using periodic boundary conditions, from twenty images recorded with the STM containing 256×256 pixels each. (x, y) represent the fast and slow scan directions, respectively. Inset: detail of the 8×8 pixels that constitute the central peak.

must be flat and (atomically) parallel to each other over an area $L \times L$ where the distance L satisfies $L/a > 2(t/a)^{1/2}$, or $L > L_0$, with $L_0 = 2(ta)^{1/2}$, and a is the lattice spacing [10].

However, experimental evidence of quantum size effects (QSE) has been found in systems that most probably *do not satisfy these criteria*. We mention just two of the most relevant:

- (a) the oscillations observed at 4 K in the derivative of the conductance versus bias voltage (d^2I/dV^2) in tunnel junctions made of thin films of Pb, Mg, Au and Ag grown onto anodized Al films deposited on a glazed ceramic substrate; the oscillations were observed up to a thickness of several tens of nanometres [11];
- (b) the oscillations of the superconducting transition temperature with film thickness observed in Sn films grown onto glazed alumina at 15 K; the oscillations persist up to a thickness of 6 nm [12].

The argument involving the conditions for subband formation in a (disordered) rough metallic surface seems related to the argument involving conditions for band formation in disordered systems, an issue that has been discussed in the literature. According to one of the axioms of solid-state theory, a sufficient condition for the formation of a band in a collection of molecules is that the system should exhibit translational symmetry. However, electrons injected into insulating liquids like liquid Ar (LAr), LKr, LXe and LCH₄ exhibit a mobility in the range several hundred to a few thousand $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ (see table 1 in Munoz's paper, reference [13]), figures which are one order of magnitude larger than the mobility of electrons in *crystalline Cu* at room temperature and which are comparable to or larger than the mobility exhibited by electrons injected into *insulating crystals* like the alkali halides at comparable temperatures (Seager and Emin, reference [13]). This indicates that translational symmetry is sufficient to warrant the formation of a conduction band, but too strong a condition to explain

the observations. The result of these experiments can be reconciled with theory if the electrons injected into these liquids are assumed to occupy *orbitals which extend over hundreds of molecules, something usually regarded as a signature of a conduction band* [13].

A similar situation arises regarding subband formation in rough metallic films. Applying the criteria of atomic parallelism to explain the observations of QSE reported in references [11] and [12] would lead to the conclusion that the metallic films studied in references [11] and [12] consist of collections of regions that are atomically parallel to the substrates over areas of the order of $A_0 = L_0 \times L_0$, $10 \text{ nm}^2 < A_0 < 100 \text{ nm}^2$. This seems possible but unlikely. The criteria of flatness and atomic parallelism may work well when the probe is a local probe as in reference [10], where QSE manifest as kinks in the conductance versus bias voltage measured with a STM in an atomically flat region of a gold film, 15 nm thick, deposited on glass at room temperature [10]. However, for experiments in which the probe involves the whole sample with an area several orders of magnitude larger than A_0 , this criterion seems sufficient (to warrant the formation of subbands) but *too strong* a condition to explain the observations reported in references [11] and [12].

The experiments reported in reference [12] indicate that the electron states of metallic films several tens of nanometres thick are still correctly described by the model of a particle in a box, in spite of the fact that the upper and lower surfaces of the sample most probably *are not atomically flat and parallel to each other*. To reconcile experiment and theory for cases in which the probe involves the whole sample, we propose a somewhat weaker criterion, that nevertheless captures the essence of the criterion set forth in reference [10] and is consistent with the argument contained in reference [12]: subbands will be observable when the r.m.s. height fluctuation $\zeta(L_0)$ satisfies $\zeta(L_0) \leq \lambda_F$, where

$$\zeta(L_0) = \sqrt{\langle [h(x, y) - \langle h(L_0) \rangle]^2 \rangle}$$

where $h(x, y)$ represents the thickness measured at position (x, y) , the symbol $\langle \rangle$ denotes an average over an area $L_0 \times L_0$ and λ_F denotes the relevant scale of distance in the problem, the Fermi wavelength (in gold, $\lambda_F = 0.52 \text{ nm}$). The lattice constant for the 70 nm thick film reported here, measured via x-ray diffraction, is $a = 0.235 \text{ nm}$; therefore $L_0 = 8.1 \text{ nm}$. We evaluated $\zeta(L)$ for the twenty images used to compute the ACF shown in figure 1, selecting for each image a submatrix of 128×128 pixels corresponding to $L = 10 \text{ nm}$. The average of $\zeta(L)$ over the twenty $10 \text{ nm} \times 10 \text{ nm}$ images is 0.45 nm, indicating that the film has a smooth texture over distances of the order of 10 nm. Within the twenty images we found two for which $\zeta(L) > 1 \text{ nm}$; these correspond to steep valleys that are remnants of grain boundaries. These valleys are separated by large distances of the order of several hundred nanometres, the typical size of the grains that coalesced to form the film. If the two images for which $\zeta(L) > 1 \text{ nm}$ are deleted, the average of $\zeta(L)$ over the remaining 18 images is 0.35 nm. These figures illustrate that the film is smooth to within one electron wavelength over distances $L \leq 10 \text{ nm}$. Consequently, the mSXW theory retains its validity.

Returning to the application of mSXW theory, the reflectivity R calculated from equations (5) and (6) and from equation (7) in reference [6] are shown in figure 2, together with the Fourier transforms $F(k_{\parallel})$ plotted as functions of the wave vector k_{\parallel} for $0 \leq k_{\parallel} \leq k_F = 12 \text{ nm}^{-1}$. For the comparison between the mSXW theory and the FS model, we have chosen the average values [$\delta = 0.689 \text{ nm}$, $\xi = 0.233 \text{ nm}$] for the exponential and [$\delta = 0.455 \text{ nm}$, $\xi = 0.480 \text{ nm}$] for the Gaussian representation of the ACF. In both cases R turns out to be a function of the angle θ that approaches zero for a certain angle, the value of which is model dependent. Because of the resemblance between the quantum theory (equation (1)) and the classical theory (equation (25) in Sondheimer's paper, reference [1]), we would expect that—for a thick rough film having many subbands, such as the gold film whose

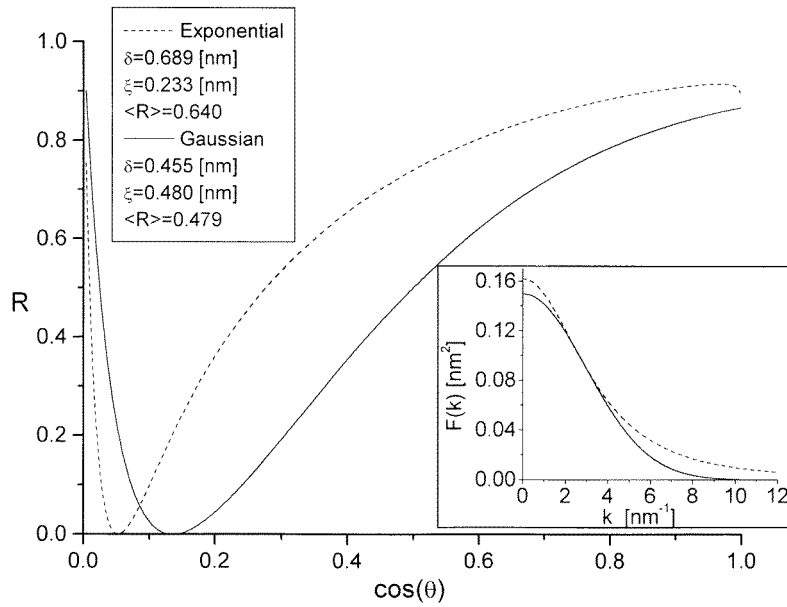


Figure 2. The reflectivity R characterizing the electron–surface scattering predicted by the mSXW theory, for a film 70 nm thick in which the average ACF is described by a Gaussian G defined by $f_G(x, y) = \delta^2 \exp[-(x^2 + y^2)/\xi^2]$, with $\delta = 0.455$ nm, $\xi = 0.480$ nm (solid curve), and by an exponential E defined by $f_E(x, y) = \delta^2 \exp[-\sqrt{x^2 + y^2}/\xi]$, with $\delta = 0.689$ nm, $\xi = 0.233$ nm (dotted curve), plotted as functions of $\cos(\theta)$. θ represents the angle of incidence between the momentum of the incoming electron and the normal to the surface. Inset: the Fourier transforms of the Gaussian $F_G(k_{\parallel}) = \pi \delta^2 \xi^2 \exp[-\xi^2 k_{\parallel}^2/4]$ (solid curve) and of the exponential $F_E(k_{\parallel}) = 2\pi \delta^2 \xi^2/[1 + \xi^2 k_{\parallel}^2]^{3/2}$ (dotted curve), plotted as functions of the wave vector $0 \leq k_{\parallel} \leq k_F = 12 \text{ nm}^{-1}$; $k_{\parallel} = k_F \sin(\theta)$.

surface roughness and conductivity were measured—the predictions of the classical and the quantum theory should be similar. This is, indeed, the case, for the discreteness arising from the quantization of k_z is washed out, in the sense that the discrete sum in equation (1) for a constant (angle-independent) R reproduces the classical Sondheimer integral with a precision of 0.5% or better.

However, since in the classical theory R is an adjustable parameter and in the quantum theory it is not, a question arises regarding which constant R should be used in the classical theory, to compare the FS model with the mSXW model. One natural way to perform such a comparison is to choose the average quantum reflectivity $R = \langle R \rangle$. For a film 70 nm thick, the mSXW theory predicts $\langle R \rangle = 0.479$ in the case of a Gaussian ACF and $\langle R \rangle = 0.640$ in the case of an exponential ACF. The values of σ/σ_0 for a film 70 nm thick predicted by FS theory, with $R = 0.479$ and $R = 0.640$, are plotted in figure 3, together with the detailed predictions of the quantum mSXW theory for a Gaussian and for an exponential representation of the ACF data. The predictions of FS theory for $R = \langle R \rangle$ coincide within 1% or better with the predictions of mSXW theory for $\ell < 50$ nm. However, as displayed in figure 3, when $\ell > t$, the increase in resistivity due to surface scattering predicted by the classical FS model for $R = \langle R \rangle$ is larger than that predicted by the mSXW theory, by an amount that substantially exceeds the level $\leq 0.5\%$ with which the sum over quantized states (equation (1)) reproduces the classical Sondheimer integral (equation (25) in reference [1]) for an angle-independent reflectivity

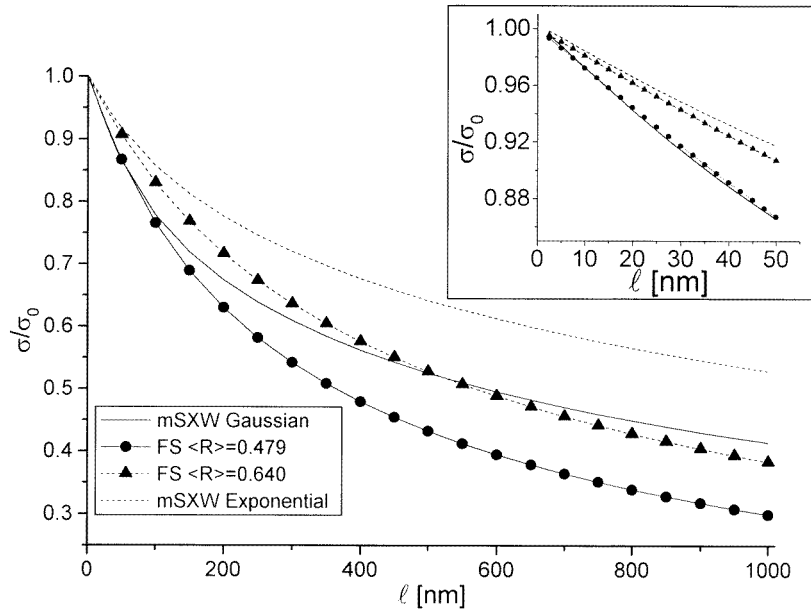


Figure 3. The film conductivity σ relative to the bulk conductivity σ_0 , as a function of the mean free path ℓ , predicted by mSXW theory, for a film 70 nm thick, characterized by an average ACF described by a Gaussian G (solid curve) and by an exponential E (broken curve). Continuous dotted curve: the prediction of the Fuchs–Sondheimer (FS) model calculated using $R = 0.479$. Triangles and broken curve: the prediction of the Fuchs–Sondheimer (FS) model calculated using $R = 0.640$. Inset: details of the prediction of mSXW theory for a Gaussian ACF (solid curve) and an exponential ACF (broken curve), the FS model for $R = 0.479$ (continuous dotted curve) and the FS model for $R = 0.640$ (triangles and broken curve), for the range $2.5 \text{ nm} \leq \ell \leq 50 \text{ nm}$.

R . The difference between the quantum prediction and its corresponding FS version grows with increasing ℓ .

The discrepancy between the classical and the quantum theories for thick films does not reside in the quantization of k_z , but arises from the fact that the quantum theory leads to a reflectivity $R[\cos(\theta)]$ that is not a constant. That the FS model is inadequate for very thin specimens is well known—because of the omission of quantum effects. The discrepancy for $\ell > t$ presented here suggests that—regardless of which model (Gaussian or exponential) is chosen to represent the ACF data—the FS model may also be inadequate for high-purity thick films, because of the omission of the angular dependence of the reflectivity R predicted by the quantum theory. These calculations also reveal that the decrease in conductivity that may be attributed to electron–surface scattering depends on which model is used to fit the same ACF data: as shown in figure 3, σ/σ_0 predicted by mSXW theory for a Gaussian representation of the data is systematically smaller than that predicted for an exponential representation of the same data. Regardless of which model is used to represent the ACF data, the results reported suggest that the relevant physical quantity controlling electron–surface scattering in continuous gold films of arbitrary thickness is the average ACF characterizing the surface of the sample on a scale of a few nm, in agreement with the accepted view regarding the conductivity of ultrathin films [4]. Such a view may have far-reaching consequences. For then, the possibility of having two metal films of different thicknesses (prepared under similar conditions of evaporation) that would exhibit the same surface reflectivity appears an odd coincidence, and the possibility of

having a family of metal films of different thicknesses *all having the same surface reflectivity* seems highly unlikely. The microscopic description provided by the mSXW theory casts doubt on one of the central assumptions that has been used to analyse conductivity data on families of metal films for many decades: the assumption that the reflectivity characterizing electron–surface scattering for a family of metal films of different thicknesses prepared under similar conditions of evaporation *is the same for all members of the family*.

To elucidate why the value of σ/σ_0 depends on the mathematical model used to represent the ACF data, in the inset of figure 2 we have plotted the Fourier transforms of the Gaussian, $F_G(k_{\parallel}) = \pi \delta^2 \xi^2 \exp[-\xi^2 k_{\parallel}^2/4]$, and of the exponential, $F_E(k_{\parallel}) = 2\pi \delta^2 \xi^2/[1 + \xi^2 k_{\parallel}^2]^{3/2}$. Comparison between F_E and F_G indicates that the transforms are similar in the intermediate region where $\xi k \sim 1$, but differ in the regions where $k\xi < 1$ and $k\xi > 1$. It has recently been suggested that if the surface roughness is a self-affine fractal, then the Fourier transform of the height–height ACF behaves as $F(k) = 2\pi \delta^2 \xi^2/[1 + A\xi^2 k^2]^{1+H}$, where A is a normalization constant and $0 \leq H \leq 1$ is the roughness exponent that measures the degree of surface irregularity [14]. The results presented suggest that the dependence of the Fourier transform $F(k_{\parallel})$ on k_{\parallel} does, indeed, have an influence on the quantum reflectivity R . The model-dependent predictions of σ/σ_0 are reminiscent of the results reported in reference [14], indicating that the conductivity limited by electron–surface scattering of a metal film bounded by a surface whose roughness is a self-affine fractal does depend on the roughness exponent H . If the fractal nature of the rough surface determines the behaviour of the Fourier transform of the ACF, then it also determines the reflectivity R and thereby the amount by which electron–surface scattering increases the resistivity of the film.

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