PHYSICAL REVIEW B **66**, 205401 (2002)

Surface-induced resistivity of thin metallic films bounded by a rough fractal surface

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(Received 20 November 2001; revised manuscript received 21 March 2002; published 6 November 2002)

We have extended the modified formalism of Sheng, Xing, and Wang [*J. Phys.: Condens. Matter* **11** L299 (1999)] to allow the calculation of the conductivity of a thin metallic film bounded by a rough fractal surface. We utilized the so-called *k*-correlation model proposed by Palasantzas and Barnas [*Phys. Rev. B* **48**, 14 472 (1993); **56**, 7726 (1997)], to describe the height-height autocorrelation function corresponding to a self-affine roughness. This extension permits the calculation of the conductivity of the film as a function of the r.m.s. roughness amplitude δ, of the lateral correlation length ξ, of the mean free path in the bulk l, and of the roughness exponent H. We found that the degree of surface irregularity, represented by the roughness exponent H characterizing the surface, does influence the conductivity of the film, as first discovered by Palasantzas and Barnas. However, this influence manifests itself for large bulk mean free paths $l \approx 1000$ nm and for large correlation lengths $\xi \approx 5$ nm, in which case the conductivity of the film for $H=1$ exceeds by about 30% the conductivity for $H=0.2$, an effect which is smaller than that reported by Palasantzas and Barnas. For correlation lengths ξ below 1 nm and mean free paths $l \approx 100$ nm, the influence of the roughness exponent H on the conductivity is reduced to below 10%, and for smaller mean free paths and correlation lengths the conductivity becomes insensitive to H. We also found that Mathiessen's rule is severily violated in the case of thin metallic films. The resistivity of the film coincides roughly with the surface-limited resistivity only in the case of ultrathin films $t<5$ nm. For thicker films 100 nm$>t>5$ nm, the resistivity of the film exceeds by some 20 to 30 % the value dictated by Mathiessen's rule. And conversely, the apparent surface-induced resistivity estimated assuming the validity of Mathiessen's rule, exceeds by nearly one order of magnitude the true surface-induced resistivity, except in the case of ultrathin films $t<5$ nm.

DOI: 10.1103/PhysRevB.66.205401 PACS number(s): 68.35.Bs, 73.50.Bk

I. INTRODUCTION

One of the fundamental problems in solid state physics that has attracted the attention of researchers for over 60 years, relates to the effect of electron-surface scattering on the transport properties of thin metallic and semiconducting films. A central issue is how the surface of the structure affects its electrical transport properties, when one or more of the dimensions characterizing the structure are comparable to or smaller than the mean free path of the charge carriers, what is known as ''size effects.''

The theoretical work concerning size effects focused for many decades on the Fuchs-Sondheimer (FS) theory, in which the electron motion is modeled by a Boltzmann transport equation (BTE) and the effect of the rough surface is incorporated into the boundary conditions that must be satisfied by the electron distribution function obeying a BTE via a specularity parameter R, that represents the fraction of electrons $0 \leqslant R \leqslant 1$ 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 where the film thickness t is much smaller than the bulk mean free path l. On the one hand, for ultra pure thin films, the conductivity of the film is expected to exhibit a stepwise increase with increasing film thickness, as a consequence of the quantization of the electronic energy levels induced by confinement of the electron gas between two parallel potential barriers, which is known as quantum size effects (QSE's). Observations consistent with these expectations have been reported in Pt films evaporated onto glass slides.[2] The modeling of electron motion via a BTE with FS boundary conditions does not account for QSE. Moreover, the resistivity of ultrathin $CoSi_2$ films observed at 4 K has been reported to increase sharply as the thickness of the film decreases below 10 nm, in a way that *cannot be accounted for by the classical model no matter what reflectivity R is used in the* FS *formalism*.[3] On the other hand, in the limit $l \rightarrow \infty$ the FS conductivity diverges as $\ln(l/t)$, implying that when the conductivity of the film is limited only by electron-surface scattering, there is no dissipation, an unphysical result that arises from the omission of quantum effects in the classical theory.

To overcome these shortcomings, a number of quantum transport theories have been published over the last two decades, that permit the calculation of the increase in resistivity arising from electron-surface scattering in a metal film bounded by a rough surface.[4–12] This work on size effects has recently been extended to compute the influence of the height-height autocorrelation function (ACF) on the conductivity of the film when the surface of the film exhibits a self-affine roughness, employing the so-called k-correlation model.[13] In addition to being of theoretical interest, the effect of a fractal surface on the surface-induced resistivity of a metal film might also be of practical and of technological interest, for the surface of gold films deposited on mica substrates under different conditions of evaporation, has been reported to conform to a self-affine model.[14]

The pioneering work of Palasantzas and Barnas (PB) (Ref. 13) utilizes the formalism developed by Fishman and Calecki,[4] where the conductivity of the film is calculated assuming that only the surface roughness contributes to electron scattering. Of course, in metallic films bulk scattering is always present. Therefore, it seems highly desirable to extend the work of PB using a theoretical formalism that allows the calculation of the resistivity of metallic films without ignoring bulk scattering. The inclusion of bulk scattering into the theoretical description of size effects arising from electron scattering by a rough surface that is self-affine, also permits an estimation of the error involved in the calculation of the surface-induced resistivity when bulk scattering is ignored.

We have recently published a modified version of the formalism of Sheng, Xing, and Wang,[11] the so-called mSXW formalism,[12] that permits the calculation of the surface-induced resistivity of thin metallic films of arbitrary thickness, characterized by a bulk mean free path l, when the height-height ACF is described by a Gaussian or by an exponential. In the present paper we present a calculation of the resistivity of a thin metallic film bounded by a self-affine rough surface, following the *k*-correlation model proposed by PB, using the mSXW formalism. The paper is organized as follows. In Sec. II we present the mSXW theory, and adapt it to include the calculation of the surface-induced resistivity when the metallic surface exhibits a self-affine roughness. In Sec. III we present the results of this work, that is, how the resistivity of the film depends on the different relevant parameters of the problem. In Sec. IV we present a discussion of the results, and a comparison between our results and those obtained previously by PB. We also present in Sec. IV an assessment of the validity of Mathiessen's rule. In Sec. V we present a summary of this work.

II. THEORY

A. Modified theory of Sheng, Xing, and Wang

Sheng, Xing, and Wang (SXW) published a calculation of size effects applicable to films of arbitrary thickness t. The calculation proceeds by computing the Green's function corresponding to a free electron gas confined between two parallel flat surfaces located at $z=0$ and $z=t$, in the absence of electron scattering in the bulk and in the absence of electron-surface scattering. The result is $VG_0(k_\parallel)=k_z\cot(tk_z)$, where $k_\parallel=(k_x,k_y)$ stands for the in-plane electron wave vector. Electron scattering is switched on in two steps. (a) First, electron scattering in the bulk is switched on, by replacing the real wave vector $k_z=\sqrt{k_F^2-k_\parallel^2}$, by the complex wave vector $q_z=\sqrt{k_z^2+i(k_F/l)}$, where k_F is the Fermi wave vector. The effect of this is to introduce a dissipation mechanism into $G_0(k_\parallel)$, for otherwise the electron states would have an infinite lifetime and the conductivity would be infinity. (b) Electron-surface scattering is switched on, and the electron self-energy $Q(k_\parallel)$ arising from scattering by the rough surface is calculated by means of the Dyson equation, leading to

$$Q(k_\parallel)=-\mathrm{Im}\int\frac{d^2q_\parallel}{(2\pi)^2}F(k_\parallel-q_\parallel)[VG(q_\parallel)], \qquad (1)$$

where Im(C) stands for the imaginary part of the complex number C; $F(k_\parallel-q_\parallel)$ is the Fourier transform of the average height-height autocorrelation function, $VG(k_\parallel)$ stands for the product of the potential V that keeps the electrons confined within the film and the Green's function $G(k_\parallel)$ describing electrons contained in the film, evaluated at either $z=0$ or $z=t$, in the limiting case $V\rightarrow\infty$.[11]

SXW use the Green's function (where k_z has been replaced by q_z), and the Kubo transport formalism to compute the reflectivity $R(k_\parallel)$ and the change of conductivity σ_F/σ_B in terms of $R(k_\parallel)$, with the result

$$R(k_\parallel)=\left(\frac{1-k_zQ(k_\parallel)}{1+k_zQ(k_\parallel)}\right)^2 \qquad (2)$$

and

$$1-\frac{\sigma_F}{\sigma_B}=\frac{3}{2}\frac{l}{t}\frac{1}{X_0N_c}\sum_{n=1}^{N_c}u_n(1-u_n^2)\times\frac{[1-R(u_n)][1-E_d(u_n)]}{1-R(u_n)E_d(u_n)}, \qquad (3)$$

with $u_n=\cos\theta_n=(n\pi)/(tk_F)$, $Xc=(tk_F)/\pi$, $N_C=\mathrm{int}(X_C)$ represents the number of occupied subbands, where int(x) stands for the integer part of x,

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

and $E_d(u_n)=\exp[-t/(u_nl)]$, which is Eq. (11) in Ref. 11. Here $\sigma_F=(\rho_F)^{-1}$ stands for the conductivity of the film, and $\sigma_B=(\rho_B)^{-1}$ stands for the conductivity of the bulk, e.g., the conductivity that would be measured *in the absence of electron-surface scattering*: $\sigma_B=[nq^2l/(\hbar k_F)]$, where n stands for the carrier concentration and q stands for the electron charge.

SXW use in their work the white noise approximation, that is, they assume that the height-height ACF $f(x,y)$ is proportional to a Dirac delta function $\delta(x,y)$, and therefore the Fourier transform $F(k_\parallel)$ is a constant. A consequence of this approximation, is the fact that the information concerning the surface roughness is incorporated into a single constant $Q(k_\parallel)=Q_0$, the self-energy of the electron gas when the ACF is described by a Dirac delta function. Although the SXW theory exhibits the distinctive feature of being the only quantum transport theory that reproduces the classical FS formalism in the case of thick and dirty films, the use of the white-noise approximation leading to the constant Q_0 severely restricts the predictive power of the theory.

To remove this limitation we have recently published a modified version of SXW theory (mSXW), where we have calculated the electron self-energy arising from electron surface scattering, for a Gaussian and for an exponential representation of the ACF, by performing the convolution indicated by Eq. (1). In the case of an exponential ACF $f(x,y)=\delta^2\exp[-\sqrt{x^2+y^2}/\xi]$, the self-energy turns out to be[12]

$$Q(k_\parallel)=\frac{2\delta^2\xi^2}{t}\sum_{n=1}^{N_c}\left(\frac{n\pi}{t}\right)^2 \times\frac{E[\pi/2,r^2(k_\parallel,q_n)]}{[1+\xi^2(k_\parallel-q_n)^2]\sqrt{1+\xi^2(k_\parallel+q_n)^2}}, \tag{4}$$

where $q_n^2=k_F^2-(n\pi/t)^2$, with $r^2(k_\parallel,q_n)=4\xi^2k_\parallel q_n/[1+\xi^2(k_\parallel+q_n)^2]$, and $E(\pi/2,r^2)$ stands for the elliptic integral of the second kind.

B. Self-affine roughness

Following PB we assume that the Fourier transform $F(k)$ of the height-height ACF corresponding to a self-affine rough surface is given by the k-correlation model[13]

$$F(k_\parallel)=\frac{2\pi\delta^2\xi^2}{(1+Ak_\parallel^2\xi^2)^{1+H}}, \tag{5}$$

where δ is the r.m.s roughness amplitude, ξ is the lateral correlation length, H is the roughness exponent related to d_f, the local fractal dimension, $d_f=3-H$, A is a normalization constant given by the self-consistent solution of

$$A=\frac{1}{2H}[1-(1+Ak_C{}^2\xi^2)^{-H}] \tag{6}$$

with $0<H\leqslant 1$, and $k_C=\pi/a_0$ is the upper cutoff wave vector in Fourier space, with a_0 denoting the distance along the (x,y) plane chosen to limit the validity of the fractal description of the surface, to account for the granularity of the atoms at short distances.[13]

Introducing Eq. (5) into Eq. (1) and using a Mittag-Leffler expansion of $VG_0(k_\parallel)=q_z\cot(tq_z)$, leads to

$$Q(k_\parallel)=-\frac{(\delta\xi)^2}{\pi t}\,\mathrm{Im}\int_0^{2\pi}d\theta \times\int_0^{k_C}\frac{q_\parallel dq_\parallel}{[1+A\xi^2(k_\parallel^2+q_\parallel^2-2k_\parallel q_\parallel\cos\theta)]^{1+H}} \times\sum\frac{\left(\frac{n\pi}{t}\right)^2}{k_F^2-q_\parallel^2-\left(\frac{n\pi}{t}\right)^2+i\frac{k_F}{l}}.$$

Performing the integration over $q_\parallel$ for a smooth function $f(q_\parallel)$ and using the approximation[12]

$$-\mathrm{Im}\int_0^{k_C}\frac{q_\parallel f(q_\parallel)dq_\parallel}{k_F^2-q_\parallel^2-\left(\frac{n\pi}{t}\right)^2+i\frac{k_F}{l}} \approx-\mathrm{Im}\int_0^{\infty}\frac{q_\parallel f(q_\parallel)dq_\parallel}{k_F^2-q_\parallel^2-\left(\frac{n\pi}{t}\right)^2+i\frac{k_F}{l}}=\frac{\pi}{2}f(q_n) \tag{7}$$

leads to

$$Q(H,k_\parallel)=\frac{2\delta^2\xi^2}{t}\sum_{n=1}^{N_c}\left(\frac{n\pi}{t}\right)^2\frac{g(H,k_\parallel,q_n)}{[1+A\xi^2(k_\parallel+q_n)^2]^{1+H}}, \tag{8}$$

where the function $g(H,k_\parallel,q_n)$ stands for the generalized elliptic integral

$$g[H,z(k_\parallel,q_n)]=\int_0^{\pi/2}\frac{d\theta}{(1-z\sin^2\theta)^{1+H}} \tag{9}$$

with $z(k_\parallel,q_n)=4A\xi^2k_\parallel q_n/[1+A\xi^2(k_\parallel+q_n)^2]$.

The function defined in Eq. (9) can be written in terms of the hypergeometric function $F(a,b,c;z)$ (Ref. 15)

$$F(a,b,c;z)=\frac{\Gamma(c)}{\Gamma(b)\Gamma(c-b)}\int_0^1 t^{-1/2}(1-t)^{c-b-1} \times(1-tz)^{-a}dt,\quad \mathrm{Re}(c)>\mathrm{Re}(b)>0. \tag{10}$$

The transformation $\sin\theta=\sqrt{t}$ applied to Eq. (9) leads to

$$Q(H,k_\parallel)=\frac{\pi\delta^2\xi^2}{t}\sum_{n=1}^{N_c}\left(\frac{n\pi}{t}\right)^2\frac{F(1+H,1/2,1;z)}{[1+A\xi^2(k_\parallel+q_n)^2]^{1+H}}. \tag{11}$$

The calculation outlined proceeds from first principles, without invoking either a classical transport equation (such as BTE) or any other quantum transport equation. It is simply based upon Kubo's linear response theory that relates the conductivity to the Green's function, and on Dyson's equation as a method of evaluating the effect of electron scattering by a rough surface through the calculation of the electron self energy arising because of electron-surface scattering. It seems appropriate to point out that the mSXW formalism outlined and the approximations on which it is based upon, have been subjected to a very stringent test. We published what we believe to be "...the first paper in which the temperature dependence and the thickness dependence of the resistivity predicted by theory, that uses as input the information contained in the surface roughness measured on a nanometric scale in an independent experiment, agrees approximately with the resistivity measured on a set of thin metallic films. *The theory contains no adjustable parameters*... ." (Ref. 16, p. 4696).

There is additional evidence that has been published supporting Kubo's linear response theory. There are two other theories that rely also on Kubo's formalism, the work of Trivedi and Aschroft (TA), Ref. 9, and the work of Tesanovic, Jaric, and Maekawa (TJM), Ref. 10. Using the surface roughness measured with a STM on a 70 nm thick gold film deposited on mica, we published an analysis of the best resistivity data available on gold films deposited on mica, using all four models: TJM, TA, mSXW Gaussian and mSXW exponential. The outcome of such an analysis is interesting: "...The first remarkable result—considering that *none* of the theories contain *any* adjustable parameter—is that all four models provide an approximate description of both the temperature and the thickness dependence of the data between 4 K and 300 K. The agreement between theory and experiment is about 15% or better in the TJM case, it is

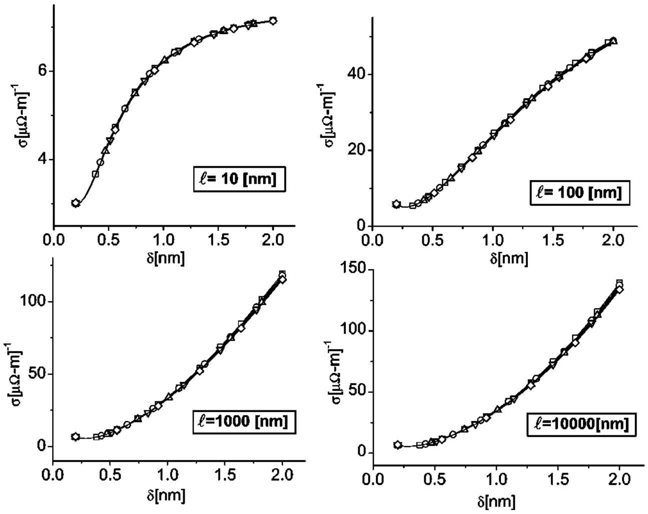

FIG. 1. Conductivity of a film 3 nm thick, characterized by a lateral correlation length $\xi = 0.2$ nm, plotted versus r.m.s. roughness amplitude δ, for different bulk mean free paths, and different roughness exponents H. Squares: $H = 0.2$; circles: $H = 0.4$; triangles: $H = 0.6$; inverted triangles: $H = 0.8$; diamonds: $H = 1.0$.

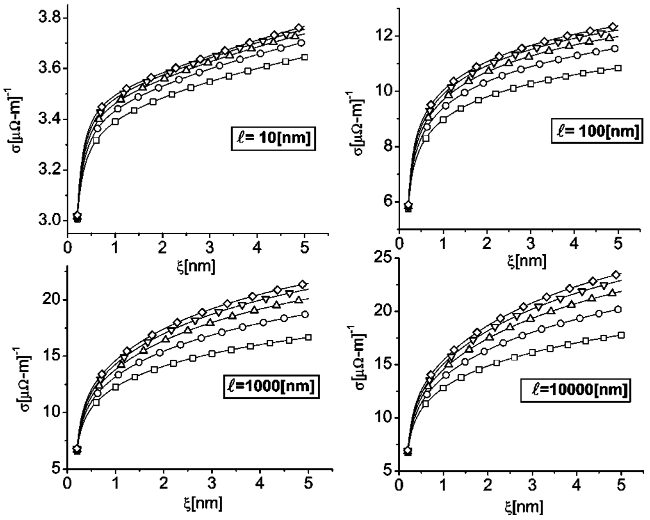

FIG. 2. Conductivity of a film 3 nm thick, characterized by a r.m.s. roughness amplitude $\delta = 0.2$ nm, plotted versus the lateral correlation length ξ, for different bulk mean free paths, and different roughness exponents H. Squares: $H = 0.2$; circles: $H = 0.4$; triangles: $H = 0.6$; inverted triangles: $H = 0.8$; diamonds: $H = 1.0$.

about 10% or better in the TA case and it is better than 7% in the mSXW case, regardless of whether we use a Gaussian or an exponential representation of the ACF.'' (p. L382, Ref. 17). That theory reproduces the experimental data to within 15% or better in the TJM case, to within 10% or better in the TA case, and to within 7% or better in the mSXW case, *without-adjustable parameters, in spite of the fact that the changes in resistivity* displayed in Fig. 6 of Ref. 16 and Fig. 1 of Ref. 17 *span roughly one order of magnitude*, constitutes quite a strong evidence that the TJM, TA, and mSXW theories based upon Kubo's linear response formalism perform very well, with the mSXW formalism performing better probably because it includes two parameters δ and ξ (instead of just one, δ) to describe the surface roughness.

Because the present extension of the mSXW theory can be considered just an extrapolation of the case where the height-height autocorrelation function is described by an exponential (which corresponds to $H = 0.5$), to the case where the autocorrelation function conforms to a self-affine roughness model, we verified that such an extrapolation reproduces the known results corresponding to the exponential autocorrelation function. We verified that in the limit $k_C \to \infty$ and $H = 0.5$, $A = 2H = 1$, the self-energy $Q(k_\parallel)$ calculated with Eq. (11), coincide with that given by Eq. (4), for the parameters H, δ, ξ, and t used in this work.

III. RESULTS

The calculations reported here were performed on the basis of Eqs. 2, 3, and 11, using the parameters corresponding to $\mathrm{CoSi_2}$. The mean free path $l = 100$ nm is that determined by Hensel and co-workers by measuring the magnetoresistance,[18] and the hole concentration $n_h = 3.0 \times 10^{28}\ \mathrm{m}^{-3}$ is that determined in an independent experiment by Badoz and co-workers by measuring the Hall effect.[19] The distance a_0 that defines the cutoff wave vector was chosen as $a_0 = a/\sqrt{2} = 0.377$ nm where $a = 0.533$ nm is the lattice constant measured on $\mathrm{CoSi_2}$.[20]

A. Influence of the r.m.s. roughness amplitude

The influence of the r.m.s. roughness amplitude δ is depicted in Fig. 1, for different roughness exponents and different bulk mean free paths, for a film $t = 3$ nm thick characterized by a lateral correlation length $\xi = 0.2$ nm. The conductivity of the film increases with increasing δ, and it increases by nearly one order of magnitude when the mean free path increases from $l = 10$ nm to $l = 100$ nm; from there on further increasing the mean free path to $l = 1000$ nm produces a further increase in conductivity of about a factor of 2, and further increasing to $l = 10\,000$ nm only increases the conductivity by about 15%. For the small correlation length $\xi = 0.2$ nm, the roughness exponent H has little effect on the conductivity of the film. The way in which the conductivity increases with increasing δ is somewhat affected by the mean free path. At $l = 10$ nm the increase in conductivity with increasing δ tends to saturate at about $\delta = 2.0$ nm; for larger l the conductivity keeps increasing with increasing δ.

B. Influence of the lateral correlation length

The influence of the lateral correlation length ξ is depicted in Fig. 2, for different roughness exponents and different bulk mean free paths, for a film $t = 3$ nm thick characterized by a r.m.s. roughness amplitude $\delta = 0.2$ nm. The conductivity of the film increases with increasing ξ, and the way in which it increases for different mean free paths is similar. When the mean free path increases from $l = 10$ nm to $l = 100$ nm, the conductivity of the film increases by about a factor of 3; from there on increasing the mean free path to $l = 1000$ nm produces a further increase in conductivity of about a factor of 2, and further increasing the mean free path to l

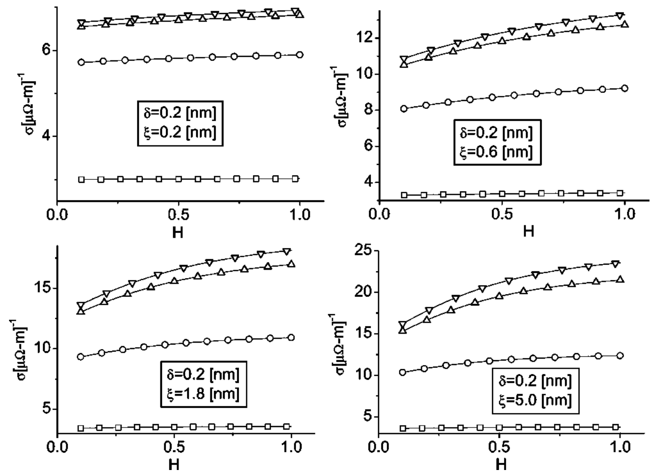

FIG. 3. Conductivity of a film 3 nm thick, characterized by a r.m.s. roughness amplitude $\delta = 0.2$ nm, plotted versus the roughness exponent H, for different bulk mean free paths, and different lateral correlation lengths ξ. Squares: $l = 10$ nm; circles: $l = 100$ nm; triangles: $l = 1000$ nm; inverted triangles: $l = 10\,000$ nm.

$= 10\,000$ nm produces a further increase in conductivity of about 10%. For small correlation lengths the roughness exponent H has little effect on the conductivity of the film. As the correlation length increases beyond $\xi = 1.0$ nm, the effect of the roughness exponent H becomes more pronounced and the conductivity of the film increases with increasing H. For large correlation lengths $\xi = 5.0$ nm and a mean free path $l = 10$ nm, the difference between the conductivity of the film for $H = 0.2$ and $H = 1.0$ is about 3%; for $l = 10\,000$ nm the difference between the conductivity of the film for $H = 0.2$ and $H = 1.0$ increases to about 30%.

C. Influence of the roughness exponent

The influence of the roughness exponent H is depicted in Fig. 3, for different different bulk mean free paths, for a film $t = 3$ nm thick characterized by a r.m.s. roughness amplitude $\delta = 0.2$ nm and different lateral correlation lengths. The conductivity of the film increases with increasing ξ and increases with increasing roughness exponent, provided that the mean free path is large. For $\delta = 0.2$ nm and $\xi = 0.2$ nm, the conductivity of the film is insensitive to the roughness exponent H, and it increases by about a factor of 2 when the mean free path increases from $l = 10$ nm to $l = 10\,000$ nm. For $\delta = 0.2$ nm and $\xi = 0.6$ nm, the conductivity of the film begins increasing with increasing roughness exponent H; the increase between $H = 0.2$ and $H = 1.0$ is about 3% for $l = 10$ nm and about 18% for $l = 10\,000$ nm. For $\delta = 0.2$ nm and $\xi = 0.6$ nm, the conductivity of the film increases by about a factor of 4 when the mean free path increases from $l = 10$ nm to $l = 10\,000$ nm. For $\delta = 0.2$ nm and $\xi = 1.8$ nm, the conductivity of the film exhibits a more pronounced increase with increasing roughness exponent H and a large mean free path; the increase in conductivity between $H = 0.2$ and $H = 1.0$ is about 3% for $l = 10$ nm and about 25% for $l = 10\,000$ nm. For $\delta = 0.2$ nm and $\xi = 1.8$ nm, the conductivity of the film increases by about a factor of 5 when the mean free path increases from $l = 10$ nm to $l = 10\,000$ nm. For $\delta = 0.2$ nm and $\xi = 5.0$ nm, the conductivity of the film exhibits yet a more pronounced increase with increasing roughness exponent H and a large mean free path; for $l = 10\,000$ nm, the increase in conductivity between $H = 0.2$ and $H = 1.0$ is about 30%. For $\delta = 0.2$ nm and $\xi = 5.0$ nm, the conductivity of the film increases by about a factor of 6 when the mean free path increases from $l = 10$ nm to $l = 10\,000$ nm.

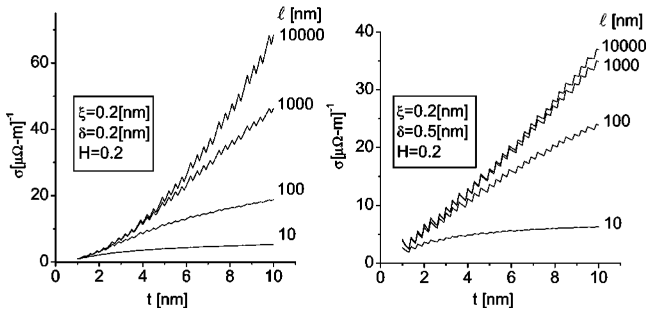

FIG. 4. Conductivity of a film characterized by a lateral correlation length $\xi = 0.2$ nm and a roughness exponent $H = 0.2$, plotted versus film thickness t, for different bulk mean free paths, and different r.m.s. roughness amplitudes δ.

D. Influence of the thickness of the film

The influence of the thickness of the film is displayed in Fig. 4, for a film characterized by a lateral correlation length $\xi = 0.2$ nm, a roughness exponent $H = 0.2$, and different mean free paths as well as different roughness amplitudes δ. The conductivity exhibits certain characteristic jumps with increasing thickness, which correspond to QSE mentioned in the Introduction, that manifest themselves as new channels of conduction opening up as the thickness increases, which translates into a new term being added to the sum in Eq. (3) each time the increase in thickness exceeds half a Fermi wavelength λ_F (in CoSi$_2$ $\lambda_F = 0.65$ nm). What is interesting is that this manifestation of QSE is severely attenuated when the mean free path is short $l = 10$ nm. However, as illustrated in Fig. 4, how much does the conductivity increase with increasing thickness, depends not only on the bulk mean free path, but depends also on how large the r.m.s. roughness amplitude δ is.

E. Influence of the cutoff wave vector

To explore the influence of the cutoff wave vector, we plot in Fig. 5 the conductivity σ_F of a film $t = 3$ nm thick characterized by $\delta = \xi = 0.5$ nm, $H = 1.0$, and a mean free path $l = 100$ nm, computed as a function of the distance a_0 chosen to define the cutoff wave vector. It is interesting to note that the distance between two neighboring atoms of Co along the (111) plane is $a/\sqrt{2} = 0.377$ nm, where $a = 0.533$ nm is the lattice constant measured on CoSi$_2$.[20] However, choosing $a_0 = 0.377$ nm leads to $k_C = 8.33\ \text{nm}^{-1}$ for the cutoff wave vector, which is smaller than the Fermi wave vector $k_F = 9.61\ \text{nm}^{-1}$ for CoSi$_2$. Therefore this crude estimation of the cutoff wave vector would lead to the exclusion of part of

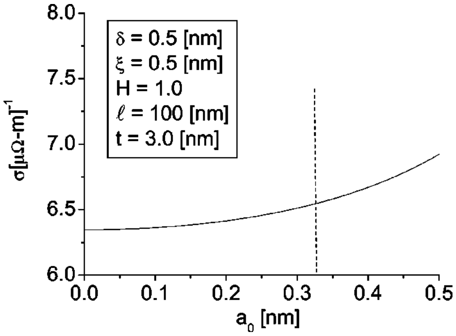

FIG. 5. Conductivity σ_F of a film $t=3.0$ nm thick plotted versus the spacing a_0 chosen to describe the granularity of the surface, for a film bounded by a rough surface that is self-affine, whose height-height autocorrelation function is described by $\delta=\xi=0.5$ nm, a roughness exponent $H=1.0$, and a bulk mean free path $l=100$ nm. The dotted vertical line corresponds to $a_F=\pi/k_F=0.327$ nm.

the occupied subbands that participate in the conduction process, something that seems difficult to justify, in spite of the fact that the roughness of the surface obviously does not conform to a fractal model, because the surface roughness at short distances is no longer random, for the granularity of the atoms becomes dominant over an atomic scale of distances. We propose instead that for metallic films, the distance chosen to define the cutoff wave vector needed to account for the granularity of the atoms that make up the surface, should be allowed to vary between $a_0=0$ corresponding to pointlike atoms, to an upper limit $a_F=\pi/k_F$, which for CoSi$_2$ corresponds to $a_F=0.327$ nm. Within these limits, increasing the cutoff spacing from 0 to a_F, induces an increase of about 3% in the conductivity of the film.

IV. DISCUSSION

A. Influence of the roughness exponent on the conductivity of the film

The results reported in this paper confirm the fact that when a thin metallic film is bounded by a rough surface that is self-affine, then the degree of surface irregularity, represented by the roughness exponent H characterizing the surface, does influence the conductivity of the film, as first discovered by PB. However, the inclusion of the bulk mean free path into the theory, seems to reduce the influence of the roughness exponent H. In fact, in agreement with the results reported by PB, the effect of the roughness exponent H on the conductivity of the film becomes more noticeable when the lateral correlation length is large, provided that the mean free path is also large. For small correlation lengths and/or small mean free paths, the roughness exponent has little influence on the conductivity of the film. In the case of large correlation lengths $\xi\approx 5$ nm and large mean free paths $l\approx 1000$ nm, the conductivity of the film for $H=1.0$ exceeds that for $H=0.2$ by about 30%, which is smaller than the effect reported by PB. There are two reasons that might explain why the influence of the roughness exponent H reported here is smaller than that reported by PB.

The first reason, is related to the predictions of the mSXW formalism concerning the scale of distances over which corrugations take place, and their relative contributions to size effects. According to arguments that have been published, the mSXW theory is able to select the scale of distance over which corrugations take place, leading to $R\approx 1$ for corrugations taking place over distances that are long when compared to λ_F, and $R<1$ for corrugations taking place over scales of distances that are comparable to λ_F to within an order of magnitude (Ref. 16, Fig. 4, p. 4692). The fact that within the theory corrugations taking place over long scales of length contribute less to size effects, is expected to lead to a film resistivity that is *less sensitive to the fractal nature of the rough surface*, than if the theory did not exhibit the ability to select scales of distances that are comparable to λ_F (to within an order of magnitude), as the corrugations that dominate size effects. However, there is a subtle point that needs to be mentioned, in connection with the electron sampling corrugations taking place over long distances.

A thin metalic film is a film such that the film thickness t is smaller than the electron mean free path l. In the case of electrons confined within a thin metalic film, for the electrons to sample corrugations taking place over long distances, comparable to the electron mean free path, the electron traveling within the film has to collide with the rough surface traveling at large angles $\pi/2>\theta>\pi/4$, where θ is the angle between the momentum of the electron approaching the rough surface and the normal to the (average) surface. But then, since the single crystal thin metalic films considered in the theory are in reality *made out of grains of finite lateral dimensions that range typically between a few nanometers and a few hundred nanometers*, electrons traveling at large angles will find a grain boundary before they have the chance "to see" a corrugation taking place over a large distance. Consequently, the resistivity of the film in this case will be dominated by grain boundary scattering rather than by the properties of the rough surface measured over very large distances, *regardless of whether the surface is fractal or not*. This has been demonstrated by van Attekum and co-workers,[21] in a nice piece of work in which the resistivity of gold films evaporated onto polished Pyrex and silicon oxide substrates *was found to decrease by about one order of magnitude upon annealing the samples*, and TEM studies of the samples proved that the decrease in resistivity upon annealing, *was directly correlated to a drastic increase of the lateral dimension characterizing the grains making up the samples (Ref. 21, Figs. 1 and 4).*

Returning to the discussion of why the present results differ from those published by PB, the second reason that could explain this discrepancy might be related to the fact that PB used a formalism proposed by Fishman and Calecki (FC) based on the Born approximation,[4] and to the fact that FC ignore the effect of bulk scattering and assume that the conductivity of the film *is limited only by electron-surface scattering*. Because of this assumption, it seems interesting to calculate the resistivity of the film $\rho_S=(\sigma_S)^{-1}$ limited only by electron-surface scattering using the mSXW formalism in the limit of large mean free paths, and to compare this resistivity with that obtained for $l=100$ nm. This compari-

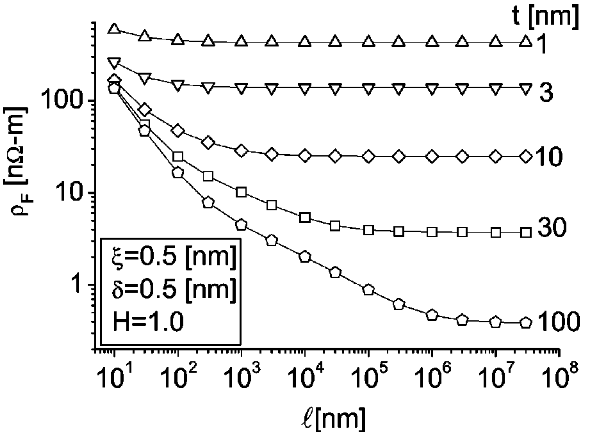

FIG. 6. Resistivity of a metallic film bounded by a rough self-affine surface characterized by $\delta = \xi = 0.5$ nm, $H = 1.0$, plotted as a function of the bulk mean free path l, for different thickness.

son will elucidate a fundamental question, whether or not Mathiessen's rule is violated, and if so, to what extent. To perform such a comparison we need to determine what a long mean free path is.

B. Influence of the bulk mean free path on the resistivity of the film

In Fig. 6 we display the resistivity of a metallic film bounded by a self affine rough surface characterized by $\delta = \xi = 0.5$ nm, $H = 1.0$ and different thickness, plotted as a function of the bulk mean free path l. From Fig. 6 it seems clear that the resistivity of the film decreases with increasing mean free path, for a mean free path of several tens of nm. As the mean free path grows larger, the resistivity of the film decreases less with increasing mean free path, until it finally levels off for large mean free paths, such that further increasing the mean free path beyond a ceratin value produces no further change in the resistivity of the film. This leveling off of the resistivity as a function of increasing mean free path defines the resistivity ρ_S of the film limited by electron-surface scattering only. From Fig. 6 it seems clear that the mean free path needed to define ρ_S is thickness dependent; it increases as the thickness of the film increases. The new and interesting result displayed in Fig. 6, is that when the film is thicker than a few tens of nm, the mean free path needed to define ρ_S reaches macroscopic dimensions. In the worst case, for a film $t = 100$ nm thick, increasing the mean free path from $l = 3.0 \times 10^7$ nm to $l = 1.0 \times 10^8$ nm produces a decrease in the resistivity of the film smaller than 0.15%. Therefore, we chose the worst case value $l = 3.0 \times 10^7$ nm to compute ρ_S for all thickness $1 \leqslant t \leqslant 100$ nm.

C. Conductivity limited by electron-surface scattering and the validity of Mathiessen's rule

One of the central issues concerning size effects relates to the resistivity ρ_S induced by electron-surface scattering in the absence of bulk scattering, and to the bulk resistivity ρ_B due to scattering in the bulk in the absence of electron-surface scattering. The question is whether the resistivity ρ_F measured on a thin film where both electron-scattering mechanisms are present (bulk scattering and surface scattering), obeys Matthiessen's rule, e.g., whether ρ_F satisfies $\rho_F = \rho_S + \rho_B$. The resistivity is proportional to the scattering rate. The additivity of the scattering rates arising from different electron-scattering mechanisms when acting together, plays an important role in solid state physics. Matthiessen's law is a powerful rule that applies to many electron-scattering mechanisms in cristalline solids: acoustic-phonon scattering, optical-phonon scattering, neutral-impurity scattering, and ionized-impurity scattering in the case of crystalline semiconductors; electron-impurity scattering, electron-electron scattering, and electron-phonon scattering in the case of crystalline metals.

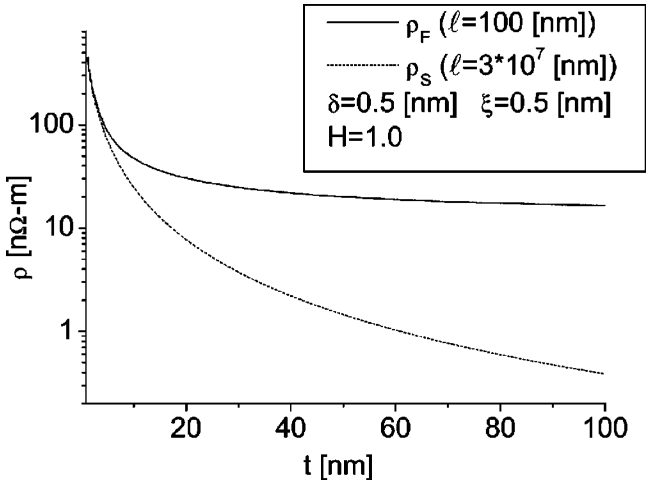

FIG. 7. Film resistivity ρ_F and surface-induced resistivity ρ_S [computed on the basis of Eqs. (2), (3), and (11)], plotted versus film thickness t, for a film bounded by a fractal surface. The height-height autocorrelation function is described by $\delta = \xi = 0.5$ nm and a roughness exponent $H = 1.0$; ρ_F was computed with $l = 100$ nm; ρ_S was computed with $l = 3.0 \times 10^7$ nm.

To address the issue concerning Mathiessen's rule in a metal film bounded by a self-affine rough surface, we plot in Fig. 7 the resistivity ρ_S arising from electron-surface scattering in the limit $l = 3.0 \times 10^7$ nm, together with the film resistivity ρ_F computed for a mean free path $l = 100$ nm, for a film whose surface is characterized by $\delta = \xi = 0.5$ nm and $H = 1.0$.

To assess the validity of Mathiessen's rule, we plot in Fig. 8(a), the quantity $\Delta\rho/\rho_F = [\rho_F - (\rho_S + \rho_B)]/\rho_F$, that ought to be zero for all thickness if Mathiessen's rule is obeyed. The *apparent* surface-induced resistivity $\rho_S' = \rho_F - \rho_B$ (e.g., the resistivity due to surface scattering that *would be estimated assuming the validity of Mathiessen's rule*) has been used in the literature as a measure of the surface-induced resistivity ρ_S.[4,22] Therefore, it seems interesting to display the ratio ρ_S/ρ_S' plotted versus film thickness (between the *true* surface-induced resistivity ρ_S and the *apparent* surface-induced resistivity ρ_S'); this is shown in Fig. 8(b). Should Mathiessen's rule hold for all thickness, then this ratio should be unity *independent of film thickness*. The degree to which the ratio ρ_S/ρ_S' departs from unity reflects the severity with which Mathiessen's rule is violated.

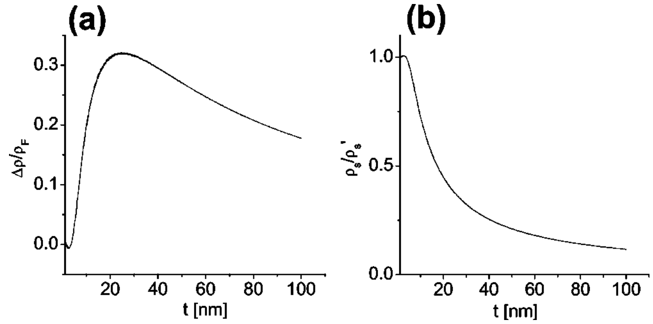

FIG. 8. (a) $\Delta\rho/\rho_F = [\rho_F - (\rho_S + \rho_B)]/\rho_F$ plotted as a function of film thickness *t*, for a film bounded by a fractal surface. The height-height autocorrelation function is described by $\delta = \xi = 0.5$ nm and a roughness exponent $H = 1.0$; ρ_F was computed with $l = 100$ nm; ρ_S was computed with $l = 3.0\times10^7$ nm. (b) Ratio between the *true* surface-induced resistivity ρ_S and the *apparent* surface-induced resistivity $\rho_S' = \rho_F - \rho_B$ plotted versus film thickness, for a film bounded by a fractal surface. The height-height autocorrelation function is described by $\delta = \xi = 0.5$ nm and a roughness exponent $H = 1.0$; ρ_F was computed with $l = 100$ nm; ρ_S was computed with $l = 3.0\times10^7$ nm.

The results displayed in Fig. 8(a) indicate that Mathiessen's rule is violated in thin metal films. For small film thickness 1.0 nm$<t<$1.2 nm, the ratio $\Delta\rho/\rho_F$ is slightly positive, of the order of $+1\%$. For 1.2 nm$<t<$3.7 nm, *the ratio* $\Delta\rho/\rho_F$ *becomes slightly negative*, of the order of -0.5%, *because the resistivity* ρ_F *of the film turns out to be somewhat smaller than* $\rho_S + \rho_B$. As the film grows thicker, deviations from Mathiessen's rule become again positive. For relatively thick films ($t = 100$ nm), the resistivity of the film exceeds by some 18% the value corresponding to the sum $\rho_S + \rho_B$. This causes the value of ρ_S' to remain finite for large thickness, whereas the true surface limited resistivity ρ_S depicted in Fig. 7 decreases rapidly with increasing film thickness. This results in a ratio ρ_S/ρ_S' that departs strongly from unity, and decreases with increasing film thickness.

The remarkable result displayed in Fig. 8 is that both *the resistivity of the film* ρ_F *and the surface-limited resistivity* ρ_S *predicted by theory are such that Mathiessen's rule is violated. For* 10 nm$<t<$100 nm, *the resistivity of the film exceeds by about 20 to 30% the sum* $\rho_S + \rho_B$. *The apparent surface-limited resistivity* ρ_S' *is not identical to* ρ_S *and, in fact,* ρ_S' *is roughly one order of magnitude larger than* ρ_S, *except in ultra thin films* $t<5$ nm.

That Mathiessen's rule is violated when the scattering mechanisms involved are electron-surface scattering and electron scattering in the bulk, has been known for over a decade.[9,10] However, to our knowledge, this paper contains the first estimation of how severe this violation is in the case of thin metallic films. It also contains a report of a *violation of Mathiessen's rule such that* $\rho_F < \rho_S + \rho_B$, *that contradicts the classical relation* $\rho_F \geqslant \rho_S + \rho_B$ demonstrated by Ziman using a variational solution of BTE.[23]

The underlying reason why Mathiessen's rule is violated seems to be related to the inherent quantum nature of electron-surface scattering. As long as electron motion is described through a classical model such as BTE, the different scattering mechanisms appearing in the collision operator in BTE might be written as the sum of the different collision operators corresponding to each scattering mechanism acting alone. The resistivity arising from each electron scattering mechanism is proportional to the matrix element representing the transition rate from the initial to the final state, and the identity of the initial and final state does not play a fundamental role, which translates into the fact that the additivity of the scattering rates leads naturally to the additivity of the corresponding resistivities, hence to the validity of Mathiessen's rule.

However, when the quantum nature of the electron scattering process becomes dominant because of the wave particle duality, as in the case of electron scattering by a rough surface, the fact that the electron is being scattered from an initial state $|\mathbf{k}\rangle$ into a final state $|\mathbf{k}'\rangle$ becomes important; consequently the identity of the initial and the final states occupied by the electron before and after the colission plays a central role in the theory. As indicated by formula (3) (regardless of which mathematical representation is chosen to describe the height-height ACE), the quantum theory leads to a resistivity that depends explicitly upon the subband index *n* that identifies the quantum states participating in the conduction process, and the total resistivity is no longer the sum of the resistivities arising from each of the different channels contributing to charge transport, *neither is the resistivity given by the sum of the different resistivities arising from each electron-scattering mechanism acting alone*. As explained in Ref. 9, because of quantum mechanics and the importance of the identity of the initial and final electron states, the additivity of the scattering rates (stemming from the statistical independence between averaging over impurities or over the phonon population, and averaging over surface roughness configurations) no longer leads to the additivity of the corresponding resistivities, hence Mathiessen's rule no longer holds.

V. SUMMARY

We have extended the modified formalism of Sheng, Xing, and Wang (mSXW, Ref. 12) to allow the calculation of the conductivity of a thin metallic film bounded by a rough self affine surface. The extension of the mSXW theory to a fractal surface allows the calculation of the conductivity of the film as a function of the r.m.s. roughness amplitude δ, of the lateral correlation length ξ, of the bulk mean free path *l*, and of the degree of surface irregularity represented by the roughness exponent *H*. We found that the roughness exponent *H* characterizing the surface does influence the conductivity of the film, as first discovered by Palasantzas and Barnas (Ref. 13). However, this influence manifests itself for large bulk mean free paths on the order of 1000 nm and for large correlation lengths on the order of 5 nm, in which case the conductivity of the film for $H = 1$ exceeds by about 30% the conductivity for $H = 0.2$. For correlation lengths below 1 nm and mean free paths of the order of 100 nm, the influence of the roughness exponent *H* on the conductivity is reduced to below 10%, and for smaller mean free paths and correlation lengths the conductivity of the film becomes insensitive to the roughness exponent H.

We also found that Mathiessen's rule is severily violated in the case of thin metallic films. On the one hand, we found that the surface-limited resistivity ρ_S plus the bulk resistivity ρ_B exceeds the value of the film resistivity ρ_F by about 1% for $1.2\ \mathrm{nm} < t < 3.7\ \mathrm{nm}$, in contradiction with the classical relation $\rho_F \geqslant \rho_S + \rho_B$ demonstrated by Ziman using a variational solution of Boltzmann transport equation. The resistivity of the film coincides roughly with the surface-limited resistivity only in the case of ultrathin films $t < 5\ \mathrm{nm}$. For thicker films $100\ \mathrm{nm} > t > 5\ \mathrm{nm}$, the resistivity of the film exceeds by some 20 to 30 % the value dictated by Mathiessen's rule. And conversely, the apparent surface-induced resistivity (estimated assuming the validity of Mathiessen's rule), exceeds by nearly one order of magitude the true surface-induced resistivity, except in the case of ultra thin films $t < 5$ nm.

ACKNOWLEDGMENTS

R.M., G.K., and L.M. gratefully acknowledge funding by FONDECYT under Contract No. 1010481. R.M. acknowledges gratefully the warm hospitality provided by J. De Hosson and the Groningen Center for Material Research, and acknowledges with pleasure illuminating and fruitful discussions with G. Palasantzas.

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