The electrical resistivity of rough thin films: A model based on electron reflection at discrete step edges

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The effect of the surface roughness on the electrical resistivity of metallic thin films is described by electron reflection at discrete step edges. A Landauer formalism for incoherent scattering leads to a parameter-free expression for the resistivity contribution from surface mound-valley undulations that is additive to the resistivity associated with bulk and surface scattering. In the classical limit where the electron reflection probability matches the ratio of the step height $h$ divided by the film thickness $d$, the additional resistivity $\Delta \rho = \sqrt{3}/2(g_0d) \times \omega / \zeta$, where $g_0$ is the specific ballistic conductance and $\omega / \zeta$ is the ratio of the root-mean-square surface roughness divided by the lateral correlation length of the surface morphology. First-principles non-equilibrium Green’s function density functional theory transport simulations on 1-nm-thick Cu(001) layers validate the model, confirming that the electron reflection probability is equal to $h/d$ and that the incoherent formalism matches the coherent scattering simulations for surface step separations $\geq 2\text{ nm}$. Experimental confirmation is done using 4.5–52 nm thick epitaxial W(001) layers, where $\omega = 0.25–1.07 \text{ nm}$ and $\zeta = 10.5–21.9 \text{ nm}$ are varied by in situ annealing. Electron transport measurements at 77 and 295 K indicate a linear relationship between $\Delta \rho$ and $\omega / \zeta d$, confirming the model predictions. The model suggests a stronger resistivity size effect than predictions of existing models by Fuchs [Math. Proc. Cambridge Philos. Soc. 34, 100 (1938)], Sondheimer [Adv. Phys. 1, 1 (1952)], Rossnagel and Kuan [J. Vac. Sci. Technol., B 22, 240 (2004)], or Namba [Jpn. J. Appl. Phys., Part 1 9, 1326 (1970)]. It provides a quantitative explanation for the empirical parameters in these models and may explain the recently reported deviations of experimental resistivity values from these models. Published by AIP Publishing. https://doi.org/10.1063/1.5020577

I. INTRODUCTION

The effect of surfaces on electron transport in thin films has attracted great interest over many decades, for both its technological importance and the underlying physics of mesoscopic systems.1–19 The Fuchs-Sondheimer (FS) model, first proposed in 1938 and extended by various researchers,20–28 is still the best known and most widely used analytical approach to describe the resistivity due to electron surface scattering. It is a classical model based on the Boltzmann transport equation, incorporating surface scattering as a boundary condition, and employing a phenomenological scattering specularity parameter $p$ which represents the probability for specular (rather than diffuse) electron reflection from a surface. Due to its simplicity and versatility, the FS model has been widely used to fit measured thin film resistivity data.6,10,24,29–31 However, the use of the single parameter $p$ to describe the electron surface scattering has resulted in ambiguity, as the understanding regarding the physical parameters that determine $p$ is limited. More specifically, some studies indicate that $p$ is affected by the surface chemistry, reporting that the surface scattering specularity of Cu(001) decreases during oxidation32 or when adding metallic cap layers,33 including Ta,34 Ni,35 or Ti.14 Some specularity is retained for insulating cap layers, which is attributed to the low surface density of states.14 Other studies primarily attribute changes in $p$ to the surface morphology, e.g., adatoms and surface vacancies, which disturb the smooth surface potential and increase the resistivity, corresponding to a scattering specularity $p = 0.34$ from measurements on evaporated silver thin films,26 and $p = 0.29$ from first principles simulations.9,36 In addition, these atoms/vacancies aggregate on the surface and form clusters which result in larger scale undulations that develop during thin film deposition and are commonly observed as surface mound features. The clusters may exhibit atomically smooth surface potentials and therefore do not alter the specularity, but nevertheless increase the resistivity by causing deviations in the thickness and scattering at discrete atomic-height surface steps.7 Consequently, many recent studies describe the effects of surface chemistry and atomic roughness with the specularity parameter $p$ within the framework of the FS model, while the scattering from larger scale undulations is commonly referred to as the surface roughness effect and is evaluated by introducing additional parameters.8,27,34 as discussed in the following paragraph.

The surface roughness of narrow conductors contributes to the resistivity increase associated with electron scattering and may be the cause for the incorrect resistivity prediction by the FS model for narrow conductors. More specifically, the reported measured resistivity of thin films $\lesssim 20 \text{ nm}$ is consistently higher than the prediction from the FS model,10,30,34,37–39 suggesting that a single parameter $p$ may be insufficient to correctly describe the resistivity vs thickness dependence. As a consequence, multiple models have been developed which...
explicitly treat surface roughness as a contributor to the resistivity. Namba\textsuperscript{23} has considered the resistivity increase due to the variation of the film thickness around its mean and derived an expression that describes the resistivity as a function of the experimentally measurable root-mean-square (RMS) surface roughness $\omega$. However, this roughness effect is secondary and the model typically underestimates the measured resistivity increase.\textsuperscript{13,30,39} Kuan et al.\textsuperscript{27,34} extended the approximate version of the FS model with an empirical factor $S$ that accounts for surface roughness effects and linearly increases the surface resistance of the FS model with an empirical factor $S = x \omega \delta^2$ and the fitting is not unique.\textsuperscript{13} When applying these FS based models to fit experimental data, the adjustable parameters raise concerns, as it is not evident whether the improved fit is the result of the correct physics or due to the larger fitting flexibility.\textsuperscript{40} Alternatively, multiple quantum mechanical\textsuperscript{3,5,41} and quasiclassical\textsuperscript{12,19,42,43} models have been developed that describe the thin film resistivity as a function of surface morphology, abandoning any adjustable parameters. These models do not include surface chemistry effects and therefore cannot completely replace the phenomenological FS model. Nevertheless, they make significant achievements towards an emerging parameter-free surface scattering model. For example, one major discovery is a $1/l^2$ dependence in the resistivity due to surface roughness without bulk scattering,\textsuperscript{3,4,19,40,41,43} which completely contradicts the zero resistivity prediction by the FS model. These quantum mechanical approaches describe the semi two-dimensional electron transport sandwiched between flat surfaces with perturbations, where the perturbations are in principle a function of the complete description of the surface structure which, however, in practice is approximated using Gaussian\textsuperscript{3} or Delta\textsuperscript{41} functions. The surface roughness is accounted for as a perturbation which is assumed to be small, that is, the surface root-mean-square roughness $\omega$ is small in comparison to the film thickness, and the lateral correlation length $\xi$ is also often approximated to be small, in comparison to the Fermi wavelength\textsuperscript{43} or the mean free path,\textsuperscript{4} in order to simplify the mathematical treatment and derive the $1/l^2$ dependency.\textsuperscript{3,4,19,41,43} Correspondingly, these quantum mechanical models describe the effect due to atomic–level roughness while neglecting larger scale roughness, i.e., surface mound features. That is, both the FS model and the more advanced quantum-mechanical treatments account for the effect of small-scale atomic-level roughness using the specularity parameter $p$ or explicit expressions, respectively, but they neglect the effect of larger-scale surface roughness on the thin film resistivity. This motivates the present investigation which focuses on exactly that: The resistivity increase associated with surface mounds of a thin film.

In this paper, we present a model that describes the effect of the surface roughness on the thin film resistivity. It uses no empirical roughness parameters but describes the resistivity contribution due to surface mounds in terms of the measurable surface morphology parameters $\omega$ and $\xi$. The model provides an additive resistivity term without accounting for atomic-level surface roughness/defects, as the latter can be accounted for with the widely used FS model or more advanced quantum mechanical approaches. The new model describes the surface roughness as an assembly of atomic-height steps which cause discrete local scattering events corresponding to finite transmission probabilities that are summed using the Landauer formalism.\textsuperscript{44,45} The resulting expression for the resistivity contains an additive term that accounts for the surface roughness effect and is independent of the specific surface scattering specularity. The derived model is validated using a combination of first-principles transport simulations on Cu layers containing atomic-height steps and electron transport measurements on epitaxial tungsten thin films with variable thickness and surface morphology.

II. MODEL FOR RESISTIVITY DUE TO SURFACE ROUGHNESS

A. Landauer formula for incoherent scattering

The Landauer formalism is a powerful and versatile approach to describe electron transport in mesoscopic systems. It was first developed in an analysis of current flow within a system with electrical field variations around localized scattering centers.\textsuperscript{44} In this analysis, the conductance due to a series of planar obstacles in a one dimensional chain is calculated by the transmission and reflection probabilities. This approach to treat the conductance as a transmission problem has been widely accepted and adapted, and was generalized to describe arbitrary scattering centers, many-channel transport, ballistic transport, and contact resistance.\textsuperscript{45–52} Within this framework, the conductance $G$, which is the inverse of the resistance $R$, of a general 1-D system can be expressed through its transmission probabilities $T_i$ of each transverse mode $i$, as\textsuperscript{48,51,53}

$$G = \frac{2e^2}{h} \sum_{i=1}^{M} T_i = \frac{2e^2}{h} MT,$$ \hspace{1cm} (1)

where $M$ is the total number of transverse modes, and $T \leq 1$ is the average transmission probability of electron waves traveling through the system. The system being studied can be either a single scattering center or a scattering region containing many scattering centers, but needs to be in reflectionless contact with two perfect conductors.\textsuperscript{48,53} A scattering-free region has a transmission probability $T = 1$ and is referred to as a ballistic conductor with a conductance $G = G_0 = g_0 A$, where $A$ is the cross-sectional area and the material property $g_0$ is the ballistic conductance per unit area. The ballistic conductance $G_0$ and its inverse, the ballistic resistance $R_0$, are independent of the length $l$ of the conductor. However, when the conductor contains a distribution of incoherent scatterers, the transmission probability decreases with increasing length, which causes the total resistance $R = R_0 + R_Q$ to increase linearly with $l$. This additional component is the usual ohmic resistance $R_Q = \rho l A$,\textsuperscript{53} where $\rho$ is the resistivity due to these distributed scatterers, while the length-independent ballistic part $R_0 = G_0^{-1}$
behaves like a contact resistance \( R_0 \). Substituting the corresponding net conductance in the form \( G = (G_0^{-1} + \rho l/A)^{-1} \) in (1) then yields

\[
\frac{1}{T} - 1 = g_0 \rho l. \tag{2}
\]

Equation (2) is valid in the ballistic regime where the conductor length \( l \) is smaller than the length \( l_{ph} \) at which energy dissipation occurs and transport becomes diffusive.\(^{40,53}\) The total transmission probability of a system with two incoherent scattering regions in series becomes\(^{46,53}\)

\[
T = T_1 T_2 + T_1 T'_2 R'_1 R_2 + T_1 T'_2 R'_2 R_2^2 + \cdots = \frac{T_1 T_2}{1 - R'_1 R_2}, \tag{3}
\]

where \( T_{1,2} \) and \( R_{1,2} = 1 - T_{1,2} \) are the transmission and reflection probabilities of regions 1 and 2, and the prime symbol denotes the reversion of the propagation direction. Rearrangement of this equation leads to \( T^{-1} = T'_1/T_1 \times (T_2^{-1} - 1) + (T_1^{-1} - 1) \), which can be immediately generalized to \( N \) scattering regions

\[
\rho_{g0l} = \frac{1}{T} - 1 = \sum_{i=1}^{N} \left[ \frac{1}{T_i} - 1 \right]. \tag{4}
\]

Microscopically scattering should be time-reversal invariant, i.e., \( T'_j = T_j \), so Eq. (4) simplifies to

\[
\rho_{g0l} = \frac{1}{T} - 1 = \sum_{i=1}^{N} \left( \frac{1}{T_i} - 1 \right). \tag{5}
\]

We note that Eq. (5) for the overall transmission probability \( T \) is derived for the case of ballistic transport, not for diffusive transport. Nevertheless, Eq. (5) is still applicable because a diffusive conductor can be divided into small ballistic segments with lengths less than the dissipation length \( l_{ph} \). The ohmic resistance of the entire conductor can then be obtained from an incoherent sum over all segments \( R_\Omega = \sum_{i=1}^{N} R_{1i} \), where the ohmic resistance of each segment \( R_{1i} = (g_0 A)^{-1} (T_i^{-1} - 1) \) is related to the well-defined transmission probability \( T_i \) of that segment, which in turn can be calculated as the \( T \) in Eq. (5). The resulting ohmic resistance in the diffusive limit \( R_\Omega = \rho l/A = (g_0 A)^{-1} \sum_{i=1}^{N} (T_i^{-1} - 1) \) then matches the ballistic expression in Eq. (5).

### B. Electron transmission at surface steps, at terraces, and in the bulk

We describe the rough surface of a thin film as a series of atomic-height steps which are separated by flat terraces, as shown in the schematic in Fig. 1. The “flat” terraces may contain adatoms, advacancies, impurities, or surface reconstructions that result in diffuse electron surface scattering, but are otherwise described by a single plane corresponding to a low index crystalline facet. Correspondingly, the sections of the thin film between the surface steps exhibit bulk and surface scattering, identical to a flat layer. In addition, however, there are discrete electron scattering events at each surface step which cause the increased resistivity due to surface roughness. We note that the scattering at the steps is assumed to be incoherent, because the carriers’ phase is randomized from bulk and surface scattering between the steps; thus, the Landauer formula for incoherent scattering can be applied. This assumption is further discussed in Sec. III.

Let us consider a film of length \( l \) with \( N \) steps and \( N \) terraces along the transport direction. Denote the terrace lengths by \( l_1, l_2, \ldots l_N \), electron transmission probabilities at each step by \( q_{1,2,\ldots N} \) and those in the terraces between steps by \( \phi_{1,2,\ldots N} \). Note however that the number of modes differs from one side of the step to the other, making transmission through the steps direction dependent, i.e., \( q_{ij} \neq q_{ji} \). More specifically, transmission through “down steps” is accompanied by a reduction of the number of transverse modes, which results in an average transmission probability smaller than unity, while travel through “up steps” causes an increase in the number of transverse modes and, in turn, nearly complete transmission as discussed in more detail below. However, an up step becomes a down step when the propagation direction is reversed. Therefore, considering the product \( \prod_j T_j/T'_j \) in Eq. (4), each reverse up step term \( q_{ij} \) is the inverse of some other forward down step term \( q_{ji} \). Correspondingly, the product becomes unity if there are equal numbers of up and down steps. In this paper, we consider conduction through “macroscopically flat” films, that is, their initial and final thicknesses are identical and their roughness is smaller than their thickness. Correspondingly, the product in Eq. (4) is unity and Eq. (5) can be applied, yielding

\[
\rho_{g0l} = \sum_{i=1}^{N} \left( \frac{1}{q_{ii}} + \frac{1}{\phi_i} - 1 \right). \tag{6}
\]

Separating the sums over \( 1/q_i \) and \( 1/\phi_i \) and expressing the contribution due to scattering between steps \( \phi_i \) in terms of the resistivity \( \rho_{ff} \) of a flat film without steps with Eq. (2) yield

\[
\rho_{g0l} = \sum_{i=1}^{N} \rho_{ff} g_0 l_i + \sum_{i=1}^{N} \left( \frac{1}{q_i} - 1 \right), \tag{7}
\]

and therefore

\[
\rho = \rho_{ff} + \frac{1}{g_0 l} \sum_{i=1}^{N} \left( \frac{1}{q_i} - 1 \right). \tag{8}
\]

This equation shows that the total resistivity of a thin film can be expressed by the sum of the resistivity \( \rho_{ff} \) of a flat
film and a term that accounts for surface roughness. Here, $\rho_{ff}$ includes bulk and surface scattering and therefore accounts for effects associated with atomic-level point defects and surface chemistry, including adatoms, vacancies, impurities, and surface reconstructions. $\rho_{ff}$ can be modeled by the Fuchs-Sondheimer model or any other model of choice. In contrast, the latter term in Eq. (8) evaluates the contribution to the resistivity from the surface roughness, which is described as a series of steps. This latter term is the primary focus of this study. To quantitatively understand this term, we note that $\rho_0 = e^2D(E_F)v_F/4$ for a Fermi liquid,52 where $D(E_F)$ is the density of states at the Fermi energy $E_F$ and $v_F$ is the corresponding average Fermi velocity. By comparing with the corresponding expression for the bulk resistivity $\rho_0^{-1} = e^2D(E_F)v_F^2\tau_0/3$, where $\tau_0$ is the bulk relaxation time, we find the relation $\rho_0 = 3/(4\rho_0v_F\tau_0)$. Using this relation, we can rearrange Eq. (8) in the absence of surface scattering ($\rho_{ff} = \rho_0$) as

$$\tau^{-1} = \tau_0^{-1} + \frac{4v_F}{3}\sum_{i=1}^{N}\left(\frac{1}{\eta_i} - 1\right).$$

(9)

where $\tau$ is the net relaxation time. This equation matches the well-known Matthiessen’s rule, indicating an additional scattering rate due to surface steps. This last term is also quite intuitive: the prefactor is the average inverse transit time of an electron through the structure (where $4/3$ appears from intuitive: the prefactor is the average inverse transit time of an electron through the structure (where 4/3 appears from

We next consider the magnitudes for the transmission probability $\eta$. For this purpose, we consider electron transport through a perfect film with flat surfaces and thickness $d$ and width $w$. It is a ballistic conductor with a conductance $G_0 = g_0dw$. Let us consider an atomically sharp down-step of height $h$, such that the film thickness is reduced to $d-h$. The overall film conductance is now limited by the number of conduction channels in the thinner section and is therefore $G_1 = g_0dw(d-h)$. This reduction of conductance from $G_0$ to $G_1$ can be interpreted as electrons being reflected at the atomic step. The electron transmission probability at this down-step becomes

$$\eta = \frac{G_1}{G_0} = 1 - \frac{h}{d},$$

(10)

where $d$ is the thickness of the source section and $h$ is the height of the down-step. Following a corresponding argument, the transmission probability for up-steps is unity, i.e., $\eta = 1$, as the overall number of available conduction channels is not affected by increasing the film thickness on one side. However, we note that any variation in potential (e.g., caused by a surface step) in a quantum mechanical system causes a non-zero reflection probability and, correspondingly, $\eta$ for an up-step is slightly less than unity. The reflection from up-steps is a local effect and therefore does not scale with $h$, as quantified by first-principles transport simulations (see also Sec. III) indicating an $h$-independent reduction in the conductance of $7 \times 10^{-5} \text{ } \Omega^{-1}$ per nm width, which corresponds to a relatively small reflection probability of, e.g., 3% or 1% for a 2 or 5 nm thick Cu layer, respectively. This reflection could be explicitly included in our model, but is neglected because (i) it is a relatively small contribution and (ii) time-reversal symmetry indicates that the total transmissions from a thin to a thick layer and a thick to a thin layer are identical. The latter point effectively means that the small reflection at an up-step equally affects the down step and, correspondingly, $\eta$ in Eq. (10) is the transmission from a pair of down and up steps. This point and particularly the validity of Eq. (10) are verified using first-principles transport simulations, as described in Sec. III.

Combining Eqs. (8) and (10) and assuming incoherent scattering from neighboring steps yield

$$\rho = \rho_{ff} + \frac{1}{2g_0d}\sum_{i=1}^{N}h_i.$$  

(11)

Here, the factor 1/2 comes from $\eta = 1$ for all up-steps, that is 50% of all steps. We focus on the case where the step-height is small in comparison to the film thickness, i.e., $h \ll d$, which leads to

$$\rho = \rho_{ff} + \frac{1}{2g_0d}\sum_{i=1}^{N}h_i.$$  

(12)

Equation (12) indicates that the resistivity contribution $\Delta \rho = \rho - \rho_{ff}$ from the surface roughness is proportional to $1/d$. Importantly, we note that the additional resistivity is only dependent on the geometry of the conductor and the material property $\rho_0$ which is easy to calculate for any metal because it depends only on the Fermi velocity and the density of states at the Fermi level, as discussed above. We demonstrate the accuracy and explore the limits of this simple expression in Sec. III using explicit first-principles transport simulations.

C. Extension to a rough 2D surface

We now extend the above expression derived for 1D transport perpendicular to discrete surface steps to a more general quasi-continuum 2D surface, represented by a surface height function $z(x,y)$, with the transport along the x axis. For the special case in which $z$ is independent of $y$, Eq. (12) for 1D transport applies and the sum $\sum_{i=1}^{N}h_i$ for discrete steps is replaced by an integral $\int_{0}^{d} |\partial z/\partial x| \cdot dx$ that describes the quasi-continuous surface height. In the more general case of randomly oriented surface steps, the steps are described as a linear combination of steps perpendicular and parallel to the transport direction, evaluated by $\partial z/\partial x$ and $\partial z/\partial y$, respectively. Scattering at the latter does not affect the electron momentum along the transport direction and is therefore neglected. We also note that the resistance caused by one single atomic step is small, so the redistribution of the current density over $y$ at different $x$ is negligible. Consequently, the film is modeled by a series of resistors in parallel, distributed over $y$, such that $\int dy/\rho = \int dy/\rho(y)$, where $\rho(y) = \rho_{ff} + (2g_0d)^{-1} \int |\partial z(x,y)/\partial x| \cdot dx$. Noting that the last term is smaller than $\rho_{ff}$, we obtain
\[
\rho = \rho_0 + \frac{1}{2\chi_0 d} \int \frac{\partial z(x,y)}{\partial x} \, dx \, dy.
\] (13)

The latter term in this expression contains no phenomenological or empirical parameter, such that the resistivity associated with the surface roughness can be directly determined from the ballistic conductance \(g_0\), the layer thickness \(d\), and the surface profile \(z(x,y)\) which is, at least in principle, directly measurable.

For the purpose of direct comparison with experiment, it is convenient to express the integral in Eq. (13) in terms of the root-mean-square (RMS) surface roughness \(\omega\) and the lateral correlation length \(\xi\) of the surface roughness. These two parameters are commonly determined when quantitatively analyzing the surface morphology of thin films, as described in more detail in Sec. IV. We consider a surface with a hexagonal close-packed array of identical conical mounts, each with a height \(H\) and a radius \(r\). For that case, Eq. (13) becomes

\[
\rho = \rho_0 + \frac{\sqrt{6} \omega}{d \chi_0 \xi}.
\] (14)

Here, the proportionality factor \(\sqrt{6}\) is determined by the geometric assumption of a hexagonal close-packed mound array and may thus be different for different surface morphologies. However, the expected variations are not large as, for example, a square mound array would lead to a correction of just 16%. More importantly, the proportionalities predicted by Eq. (14) are independent of the surface morphology model. In particular, the additional resistivity \(\Delta \rho\) due to the surface roughness is inversely proportional to the layer thickness and the specific ballistic conductance, and proportional to the ratio \(\omega \xi / \xi\), which corresponds to the average surface slope.

III. FIRST-PRINCIPLES TRANSPORT SIMULATIONS

Non-equilibrium Green’s function (NEGF) density functional theory (DFT) calculations are performed to confirm the above-described step-model, to verify the step transmission probability \(\eta\) for an actual atomistic system, and to investigate possible effects from coherent scattering at neighboring steps. The simulations are done using as a material system a 6 monolayer (ML) thick Cu(001) layer, which was primarily chosen because of the relatively simple Cu Fermi surface and the s-character of the conduction band that provides an accurate electronic structure with a limited-size localized atomic orbital basis set. A scattering region is placed between two electrodes that are semi-infinite along the [100] transport direction. Both the scattering region and the electrodes have a height of 6 ML corresponding to \(3a = 11\) Å where \(a = 3.615\) Å is the experimental room-temperature Cu lattice constant, and a width of \(a\) along [010], but with periodic boundary conditions such that they represent a thin film that is infinite in the [010] direction. The corresponding supercell size is \((L + 1.5a) \times a \times 10a\), where \(L = (1-17)a\) is the length of the scattering region, \(1.5a\) on each side of the scattering region corresponds to 3 ML of each electrode that is included as a buffer layer, and \(10a\) along [001] provides space for the six ML of the layer plus a vacuum of \(7a\) above/below the Cu layer. The electronic structure is calculated with SIESTA, using a \(\Gamma\)-centered \(12 \times 18 \times 1\) \(k\)-point mesh for the electrodes, and a \(1 \times 18 \times 1\) mesh for the scattering region. All calculations use single-zeta basis with polarization orbitals, an energy shift of 0.02 Ry, a norm-conserving pseudopotential for copper that includes all core electrons up to the 3p electrons, and the Perdew-Burke-Ernzerhof (PBE) exchange correlation functional. Electron smearing is carried out with a Fermi-Dirac occupation function with a temperature of 100 K. The electronic transport properties are calculated using the TRANSIESTA code with zero bias. Green's functions of the electrodes are determined with 32 points on the complex contour. After the TRANSIESTA calculations, the transport coefficients are calculated with a \(1 \times 255 \times 1\) \(k\)-point mesh. All computational parameters, including the \(k\)-point mesh density, basis set, energy shift, and points on the complex contour, are chosen such that the presented transmission probabilities are numerically converged to within 1%. The calculated ballistic conductance for the 6 ML thin film along [100] is \(0.996 \times 10^{15} \Omega^{-1} \text{m}^{-2}\). This value is 7% smaller than the ballistic conductance for bulk Cu, \(g_0 = 1.08 \times 10^{15} \Omega^{-1} \text{m}^{-2}\), calculated using a comparable computational parameter set. The latter value compares well to a reported \(g_0 = 1.10 \times 10^{15} \Omega^{-1} \text{m}^{-2}\) based on the Fermi velocity and density of states of bulk Cu, consistent with the overall computational accuracy of \(\pm 1\%\).

The electron transmission probability at a surface step is calculated using a scattering region where the left and right sides of the thin film are misaligned, forming an up-step on the top surface and a down-step at the bottom surface, as illustrated with a schematic in Fig. 2. This is done by raising the right half of the scattering region as well as the right electrode by an integer number of monolayers (ML), and correcting for the stacking sequence such that all Cu atoms both in the scattering region and in the two electrodes occupy sites of the same fcc lattice. This geometry with both an up-step and a down-step on opposite surfaces is chosen because it removes any ambiguity associated with defining the thickness, as the thickness remains constant throughout the simulated configuration. In addition, including an up-step within the calculation of a down-step corrects to first order the approximation of \(\eta = 1\) for up-steps, such that the model presented in Sec. II should become valid even for the case of very thin layers where the reflection at up-steps is not negligible. We note that this geometry illustrated in the inset of Fig. 2 is designed to quantify the scattering at a local surface step, while the overall rough surface that is considered in the model in Sec. II is represented in Fig. 1. The conductance is...
than the nominal thickness $h$, confirming our assumptions that lead to the simple pre-
gobtained using a linear curve fitting with a y-intercept fixed at $d = 7.25$ ML. The inset illustrates the simulated structure.

calculated as a function of step height $h$, from $h = 0$ ML for a
perfectly smooth layer to $h = 1$ ML for a single monolayer
step to $h = 6$ ML for a completely misaligned thin film to $h = 9$ ML for a discontinuous film.

Figure 2 is a plot of the calculated conductance per area $g(h)$, in units of $\Omega^{-1} m^{-2}$, as a function of the step height $h$, for the simulated 6 ML-thick Cu (001) layer. The right y-axis shows the corresponding transmission probability $\eta(h)$, which is determined from the ratio of the calculated conductance $g(h)$ divided by the conductance of the perfectly smooth ($h = 0$) film, where the latter has a $g(h = 0) = 0.996 \times 10^{15}$ $\Omega^{-1} m^{-2}$. Introducing a single atomic-height step ($h = 1$ ML) results in a reduction of $g$ by 17% to $0.825 \times 10^{15}$ $\Omega^{-1} m^{-2}$, corresponding to $\eta = 0.829$. $g$ decreases with increasing step height, reaching $0.189 \times 10^{15}$ $\Omega^{-1} m^{-2}$ for $h = 6$ ML. This latter case ($h = 6$ ML) corresponds to the two sides of the thin film being nominally completely misaligned, such that the conductance should (classically) be zero. In contrast, the calculated $\eta = 18.9\%$, which is attributed to tunneling that is facilitated by the wave function extending well beyond an inter-planar spacing above at the Cu(001) surface. In fact, even $h = 7$ ML leads to a finite $\eta = 0.045$, while $h \geq 8$ ML results in a negligible $\eta < 0.0003$. The calculated $g(h)$ data exhibit an approximately linear dependency, as illustrated by the dashed line through the data points in Fig. 2, which is obtained using a linear curve fitting with a y-intercept fixed at $\eta = 1$. This linear relationship is in perfect agreement with Eq. (10), confirming our assumptions that lead to the simple prediction of the electron transmission probability at a step $\eta = 1 - \frac{\hbar d}{\hbar d}$. We note, however, that the x-axis intercept indicates an effective thickness $d = 7.25$ ML which is considerably larger than the nominal thickness $d_0 = 6$ ML. This difference is again attributed to the wavefunction extending well ($\sim 1$ ML) above the surface. This difference of the order of one ML becomes negligible for larger (experimentally more realistic) layer thicknesses.

Figure 3 is a plot of the calculated total transmission probability $T$ of two-step structures which are investigated to quantify the possible correlation between neighboring steps. The simulated structures exhibit two steps which are each one monolayer high ($h = 1$ ML) and are separated by a distance $l$, as illustrated in the schematics in the inset. As before (in Fig. 2), each step is formed by the misalignment of a 6-ML-thick film, and therefore corresponds to a pair of an up-step and a down-step on opposite surfaces. Two configurations are investigated, namely, a structure where the two mismatch junctions are in the same direction, as illustrated in Fig. 3 in blue, and a structure where the mismatch of the two junctions is in the opposite direction, as illustrated in red. The corresponding data points are plotted as blue x’s and red circles, respectively, showing the calculated total transmission probability $T$ through the two junctions as a function of their separation $l$. If the steps are separated by only a single lattice constant, $l = a = 3.615$ Å, the calculated transmission probability is 0.774 and 0.758 for the blue and red configurations, respectively. Increasing $l$ results in a decrease in the transmission, which converges to $T = 0.74 \pm 0.01$ for $l \geq 6a$, as indicated by the dashed horizontal line in Fig. 3. We attribute the higher $T$ at small $l$ to direct tunneling across the terrace between the step-junctions. The transmission probability exhibits some oscillations even for $l > 6a$, which is attributed to coherent scattering at the two step junctions. The positions of the blue maxima match the red minima and vice versa, with an oscillation wavelength that increases with $l$ and reaches $\sim 2$ nm for $l > 10a$. This is considerably larger than the bulk Cu Fermi wavelength of 4.6 Å, suggesting that the oscillations are attributed to standing waves within the terrace with length $l$. We note that coherent scattering within these structures is possible because these calculations are done without any incoherent scattering centers, i.e., there are no phonons, point defects, or surface irregularities on the terrace. In contrast, Sec. II assumes the presence of incoherent scattering events, which is a prerequisite for the validity of Eq. (5) and subsequent derivations. To illustrate this point, we apply the incoherent formism to the two-step problem. Equation (3) for incoherent scattering simplifies for the case of two identical and symmetric steps to $T = \eta/(2 - \eta)$, where $\eta$ is the transmission probability.
at a single step. There are two ways to extract \( \eta \) from the data presented in Fig. 2. Direct calculation of a 1-ML-step yields \( \eta = 0.829 \), while the linear fit \( \eta = 1 - h/d \) with \( h/d = 1/7.25 \) yields \( \eta = 0.862 \). Both values have merits: the former corresponds to the actual 1-ML-high step used in the step-interaction calculations presented in Fig. 3, while the latter value has a considerably smaller computational uncertainty as it is obtained from multiple calculated configurations. These multiple configurations also reduce “noise” associated with specific ratios of the electron wavelength with a particular step height or thin film thickness, which reduces the uncertainty of the latter value. Correspondingly, we use the two values to define a range \( \eta = 0.829–0.862 \) for the transmission probability for a 1-ML-high step, and calculate the corresponding transmission across two sequential incoherent 1-ML-high steps to be \( T = 0.708–0.757 \). This range corresponds to \( T = 0.73 \pm 0.02 \) for the incoherent prediction, which is in excellent agreement with the calculated \( T = 0.74 \), indicating that the incoherent scattering formalism is applicable even for this somewhat extreme case of completely coherent scattering at two neighboring steps of a very thin (6 ML = 11 Å) film. We note that the 2.7% uncertainty in the incoherent prediction \( T = 0.73 \pm 0.02 \) leads to a larger relative uncertainty of 10% in the predicted resistivity, since according to Eq. (2) the resistivity is proportional to \( (1/T-1) \). However, for the case of a more realistic system which includes incoherent scattering both in the bulk and at the terrace surfaces, the incoherent formalism becomes increasingly more valid which is expected to reduce both the uncertainty in the predicted \( T \) and any possible deviation between \( T \) for coherent and incoherent scattering.

In summary, the results in Figs. 2 and 3 validate the model presented in Sec. II. In particular, they support (i) the assumption that the transmission probability of a surface step can be accurately approximated from the ratio of the step height over the layer thickness, as defined in Eq. (10), and (ii) the assumption that scattering at neighboring steps is well described within an incoherent scattering formalism.

**IV. EXPERIMENTAL VALIDATION**

Transport measurements were performed in order to evaluate the prediction of Eq. (14) for the thin film resistivity as a function of the root mean square surface roughness \( \omega \) and the lateral correlation length \( \xi \) of the surface morphology. We have chosen as an experimental model system epitaxial W(001) films, primarily because the high melting point facilitates epitaxial (single-crystal) growth of thin continuous layers on insulating substrates down to thicknesses of 4 nm.62,63 and we have previously developed in situ annealing procedures that allow variations in the surface roughness with negligible changes in the crystalline quality.39 4.5–52 nm thick W(001) films were deposited on MgO(001) substrates in a three chamber ultrahigh vacuum DC magnetron sputter deposition system with a base pressure <10\(^{-9}\)Torr following the procedure in Ref. 62. After deposition, they were transported without breaking vacuum to the analysis chamber maintained at a base pressure of 10\(^{-9}\)Torr for in situ resistivity measurements by a linear 4-point probe, as described in Ref. 35. In order to vary the surface morphology, some of the samples were annealed at a base pressure <10\(^{-9}\)Torr at 1000°C for 2 h followed by another in situ resistivity measurement. After the samples were removed from the vacuum, they were dropped into liquid N\(_2\) within 2 s, followed by 4-point-probe measurements at 77 K in liquid N\(_2\). The layer thickness and surface roughness were determined by x-ray reflectivity (XRR) analyses according to the procedure described in Ref. 14. Extensive structural characterization was done to characterize the layer microstructure by various x-ray diffraction methods similar to the procedures described in Refs. 62,64, and 65 for epitaxial ScN(001), Sc\(_{1-x}\)Al\(_x\)N(001), and W(001) layers, confirming that there is no significant change in the structural quality of the samples during annealing for \( d \leq 20 \) nm, as previously reported in Ref. 62. The surface morphology is quantitatively analyzed by extracting the height-height correlation function \( H(r) \) from atomic force micrographs (AFM) following the procedure described in Refs. 66 and 67. The RMS surface roughness \( \omega \) and the lateral correlation length \( \xi \) are then obtained by fitting of the \( H(r) \) data assuming a self-affine surface morphology68 using eight micrographs for each sample, as previously reported in Ref. 39. A total of nine W(001) layers with thickness \( d = 4.5–52 \) nm are investigated, yielding a range of surface morphologies with \( \omega = 0.25–1.07 \) nm and \( \xi = 10.5–21.9 \) nm. Annealing causes a reduction in \( \omega \), for example, from 0.67 ± 0.05 nm for \( d = 5 \) nm to 0.29 ± 0.03 nm for \( d = 4.5 \) nm or from 1.07 ± 0.11 nm for \( d = 48 \) nm to 0.25 ± 0.03 nm for \( d = 52 \) nm, while \( \xi \) remains nearly constant at 13 and 20 nm, respectively. That is, annealing considerably reduces \( \omega \) (by a factor of 2–4) without affecting \( \xi \) (which increases by 6%–14%).39 This provides the ideal sample set to test the presented model, which predicts a resistivity change as a function of the ratio \( \omega/\xi \).

To check the validity of our model, we compare the measured resistivity with the prediction from Eq. (14). For this purpose, the flat film resistivity \( \rho_{ff} \) is set equal to the expected \( \rho_{FS} \) calculated from the classical Fuchs–Sondheimer model:

\[
\rho_{FS} = \rho_{bulk} \left[ 1 + \frac{3}{8} (1-p) \frac{\lambda}{d} \right].
\]

(15)

where \( \lambda = 15.5 \) nm is the reported bulk mean free path at room temperature,69 \( p \) is the specularity parameter of the two surfaces which is set to zero (completely diffuse scattering) according to Ref. 63, and the bulk resistivity \( \rho_{bulk} = 5.6 \) and 1.08 \( \mu \)Ω cm at 295 and 77 K, respectively, is obtained from the measured resistivity of thick samples (320 and 390 nm) which is corrected for by the relatively small 1.8% and 8.3% resistivity size effect in these samples at 295 and 77 K. The mean free path at 77 K, \( \lambda = 80.4 \) nm, is determined from the room temperature mean free path and the fact that the product \( \lambda \times \rho_{bulk} \) is temperature independent. The flat film resistivity determined with Eq. (15) is significantly smaller than the measured total resistivity \( \rho \). We attribute the difference \( \Delta \rho = \rho - \rho_{ff} \) to the surface roughness effect. It corresponds to the measured resistivity contribution due to surface
roughness and is plotted in Fig. 4 as a function of \( \omega / (\xi d) \), where all three values, \( \omega, \xi, \) and \( d \), are directly obtained from AFM and XRR measurements. The plot includes \( \Delta \rho \) of the as-deposited and annealed W(001) layers denoted by triangles and squares, respectively, measured at two temperatures, 77 K and 295 K, as indicated by the blue and red color. The dashed lines are the result from linear curve fitting through the origin, which is done independently for 77 and 295 K. The measured data are well described by the dashed lines suggesting that \( \Delta \rho \) is proportional to \( \omega / (\xi d) \), as predicted by Eq. (14). That is, our measured resistivity data support the model developed in Sec. II. We note that the thinnest samples have the largest experimental uncertainty in \( \Delta \rho \), as indicated by the plotted error bars in Fig. 4. This may, in turn, explain the relatively large scatter from the linear trend for the data points at \( \omega / (\xi d) = 0.0049 \), which are obtained from the thinnest sample with \( d = 4.5 \) nm. The fitting provides values for the slopes of \((8.3 \pm 0.8) \times 10^{-15} \, \Omega \, \text{m}^2\) and \((11.0 \pm 1.2) \times 10^{-15} \, \Omega \, \text{m}^2\), for 77 and 295 K, respectively, from which a specific ballistic conductance of \((1.5 \pm 0.2) \times 10^{14} \, \Omega^{-1} \, \text{m}^{-2}\) and \((1.1 \pm 0.1) \times 10^{14} \, \Omega^{-1} \, \text{m}^{-2}\) can be determined by directly applying Eq. (14) while neglecting the substrate-layer roughness. We note that the plotted \( \Delta \rho \) is systematically lower at 77 K than at 295 K, corresponding to a 24% lower slope. This observation deviates from our model that predicts a temperature independent uncertainty. We attribute this deviation to the uncertainty in the determination of \( \rho_{ff} \), which requires values for the bulk resistivity and the bulk electron mean free path as shown in Eq. (15). In particular, a small error in the low-temperature bulk resistivity due to residual impurities leads to a systematic error in \( \rho_{ff} \) that could explain the 24% difference in the slope. More importantly, the exact slope is affected by the value of the bulk mean free path, which is taken as \( \lambda = 15.5 \) nm from Ref. 69. However, there is no consensus in the literature regarding the most appropriate value for \( \lambda \) of W. For example, fitting our data with another reported \( \lambda = 38 \) nm which corresponds to a product \( \lambda \times \rho_{bulk} = 2.1 \times 10^{-15} \, \Omega \, \text{m}^2\) yields a specific ballistic conductance of \(9.88 \times 10^{14} \, \Omega^{-1} \, \text{m}^{-2}\). This value is close to the reported first-principles predictions for the ballistic conductance for W along the [100] and [110] transport directions of \(9.5 \times 10^{14}\) and \(8.7 \times 10^{14} \, \Omega^{-1} \, \text{m}^{-2}\), respectively. We emphasize here again that the primary conclusion from the experimental measurements is the linear relationship in Fig. 4 that confirms our new model and particularly the resistivity prediction of Eq. (14) which includes no phenomenological or empirical parameters.

V. DISCUSSION

In this paper, we have presented in Sec. II a new explicit expression for the effect of surface roughness on the resistivity of a thin film, have provided in Sec. III first-principles calculations that validate the assumptions for incoherent scattering and the electron reflection probability at surface steps used in the development of the new model, and have presented in Sec. IV results from experimental transport measurements that support the derived expression for the resistivity. The primary focus of this Sec. V is to discuss how the new derived expression relates to existing models for the resistivity of thin films.

The exact expression of the surface roughness effect from our model is presented in Eq. (13), which expresses the resistivity due to surface roughness in terms of the average resistivity of the film, \( \rho_f \), and the average resistivity of the flat surface, \( \rho_{ff} \). This equation is valid for any type of surface roughness and can be applied to both clean surfaces and surfaces with contaminants. The model is based on a combination of the Fuchs-Sondheimer model and the 2D electron gas model, and it accounts for the effects of surface roughness on the electron scattering in the film. The model is valid for both clean surfaces and surfaces with contaminants, and it can be applied to a wide range of surface roughness levels.

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![Fig. 4. The difference \( \Delta \rho \) between the measured resistivity and the Fuchs-Sondheimer prediction, for the as-deposited (triangles) and annealed (squares) W(001) layers measured at 77 K (red) and 295 K (blue), plotted against the measured \( \omega / (\xi d) \). The dashed lines are the result from linear fitting.](image-url)
within the conventional FS model to an increase in diffuse surface scattering and therefore a reduction in the phenomenological specularity parameter $p$. That is, previous studies that determined the surface scattering specularity by fitting measured resistivity data with the FS model may have reported a $p$-value that is lowered by the surface roughness. More problematic is the fact that a surface that causes completely diffuse surface scattering ($p_s = 0$) but also exhibits some roughness ($\Delta p < 0$) yields resistivity vs $d$ data that corresponds to a negative $p$, which is outside the limits $0 \leq p \leq 1$ of the FS model. This may explain the reported discrepancy between measured thin film resistivities and the FS model prediction.\textsuperscript{30,34,37,38}

A well-known extension to the FS model which has the goal to resolve this discrepancy has been proposed by Kuan et al.,\textsuperscript{27,34} who introduced an empirical parameter $S$ to account for surface roughness effects such that the FS prediction becomes

$$\rho = \rho_{\text{bulk}} \left[ 1 + \frac{3}{8} \left( 1 - p \right) \frac{\lambda}{d} S \right]. \quad (18)$$

In this expression, the contribution from surface roughness on the resistivity is dependent on the surface scattering specularity in such a way that the effect of roughness becomes negligible for a surface with a completely specular ($p = 1$) scattering. In contrast, the surface roughness contribution in our model is an additive term which is independent of the surface specularity. This becomes evident when setting Eqs. (14) and (18) equal, applying $\rho_{ff} = \rho_{\text{bulk}} [1 + 3(1 - p)\lambda/8d]$ and solving for the roughness factor $S$, which is explicitly expressed based on our model by

$$S = 1 + 4 \sqrt{\frac{2}{3}} \frac{\omega}{\xi} \frac{1}{1 - p}. \quad (19)$$

This indicates that $S$ is a direct function of the surface scattering specularity and diverges for completely specular surface scattering.

An earlier attempt to account for the surface roughness effect was made by Namba,\textsuperscript{23} who considered the effect of variations in the film thickness on the resistivity. More specifically, the (rough) thin film resistivity is expressed as an integral over the flat film resistivity $\rho_p(d + \Delta d)$ where $\Delta d$ is the deviation from the average thickness $d$ and varies with position $x$. Applying the FS model to the Namba approach and assuming that the RMS roughness $\omega$ is much smaller than $d$, we derive

$$\rho = \rho_{ff} + \frac{3}{8} \left( 1 - p \right) \rho_{\text{bulk}} \frac{\lambda \omega^2}{d^2}, \quad (20)$$

where $\rho_{ff} = \rho_{\text{bulk}} [1 + 3(1 - p)\lambda/8d]$ is the flat film resistivity for a constant $d$. Thus, the second term in Eq. (20) corresponds to the resistivity contribution from the surface roughness according to Namba. This term is proportional to $(1/d)(\omega^2/d^2)$, while our model in Eq. (14) predicts a $(1/d)(\omega/\xi)$ proportionality. Since $\omega/d$ is small, the roughness effect described by Namba is only a secondary effect in comparison to our model. This is because, contrary to our model, the surface roughness in the Namba model does not cause additional electron scattering, such that the resistivity becomes independent of the lateral correlation length $\xi$. This is in clear contradiction to our expression which predicts a considerably stronger impact of the surface roughness on the thin film resistivity.

We note that existing quantum models\textsuperscript{3,4,41,43} also provide expressions for the resistivity as a function of surface morphology. However, as summarized in the Introduction section, these models focus on the effect of small-scale atomic-level roughness while neglecting the effect of larger-scale surface roughness which is the focus of our study. Therefore, these models provide expressions for $\rho_{ff}$, which is within this paper defined as a “known” resistivity of a flat surface that contains atomic-level roughness and can be described with the FS model or these more advanced quantum models. Correspondingly, these models are compatible with our derivation for the effect of surface roughness but cannot be directly compared with it since they focus on a distinctly different effect which results, within our framework, in an additive resistivity term.

In summary, in this section we have directly compared our expression for the resistivity due to surface roughness with predictions from existing models. More specifically, we find that (i) surface roughness causes a decrease in the apparent specularity parameter $p$ within the FS model, which may become negative and, thus, unphysical, (ii) the roughness factor $S$ within the model of Kuan et al. is a function of $\omega/\xi$, which is consistent with our prediction, but $S$ tends to infinity for specular surface scattering, suggesting a possible divergence of this model with our prediction for $p \approx 1$, (iii) the Namba model predicts a resistivity due to roughness which is independent of the lateral length scale for roughness and is relatively small in comparison to our prediction, and (iv) existing quantum models complement our model by providing expressions for $\rho_{ff}$.

VI. CONCLUSIONS

We have presented a new model that predicts the impact of surface roughness on the resistivity of a thin film. This is done by analyzing electron scattering at discrete surface step edges. This resistivity is found to be additive to the flat film resistivity, and can be expressed in terms of a transmission probability at each step by applying a Landauer formalism. The transmission probability at a single step decreases linearly with the step height, as confirmed with NEGF-DFT simulations of six-monolayer-thick Cu(001) layers. The simulations also show that the transmission across a two-step structure converges to the expected incoherent transmission across two individual steps with increasing step-step separation, indicating that the incoherent scattering approximation is applicable to layers with reasonably flat surfaces. Generalizing the transport model to 2D surfaces yields a parameter-free expression [Eq. (13)] for the resistivity due to surface roughness. It is proportional to the average surface slope and inversely proportional to the film thickness and can be expressed, for direct comparison with experiments, in terms of the RMS surface roughness $\omega$ and lateral correlation...
length $\xi$, showing a linear dependence $\Delta \rho \sim \omega l(\xi d)$. Experimental validation of this proportionality is done by measurements of the surface morphology and resistivity of the annealed and as-deposited epitaxial W(001) films. They reveal a significant resistivity contribution $\Delta \rho$ due to the surface roughness ranging from 0.3 to 11.2 µm. Plotting the measured $\Delta \rho$ at both 295 and 77 K against the measured $\omega l(\xi d)$ suggests a linear relationship, confirming the model’s prediction. Finally, our result is compared to previously developed surface roughness models, providing a quantitative explanation of their empirical parameters as well as revealing the limitations of existing models. This work provides a parameter-free method to evaluate the surface roughness contribution to the thin film resistivity.

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