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Influence of anomalous roughness growth on the electrical conductivity of thin films

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In this work it is explored how anomalies in the dynamic evolution of self-affine interface roughness influence electrical conduction in thin films. For metallic films if the roughness amplitude w increases faster than the correlation length ξ with increasing film thickness that leads to higher scattering and thus lower conductivity. The latter still increases with increasing roughness exponent H due to interminiband scattering. The opposite behavior was observed for semiconducting films where only one miniband was occupied, therefore excluding interminiband scattering effects. Therefore, dynamic roughness evolution and interminiband scattering can interact and influence the thickness dependent conductivity of thin films. Indeed for semiconducting films the evolution of the long wavelength roughness parameters w and ξ can obscure the smoothing effect due to increment of the roughness exponent H and decrease the conductivity in the case of rapid surface/interface roughening.

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I. Introduction

Surface/interface roughness in thin films strongly influences their electrical conductivity since it induces additional charge scattering.^{1–20} Indeed, scattering by random roughness alters the size and shape of quantum size effects (QSE) in a manner that depends strongly on the nature of the roughness correlation function.^{14,18} Besides metallic films, for quantum wells it has been shown that the mobility of the two-dimensional electrons in modulated-doped AlAs/GaAs well is affected by interface roughness.⁸ Moreover, the agreement between theoretical and experimental data for the Hall mobility of metal-oxide-semiconductor field effect transistors⁹ (MOSFETs) and for the electron mobility of Si inversion layers¹⁰ improves by assuming an exponential correlation function for the Si/SiO₂ interface. Interface roughness also affects electron subbands of, e.g., InAs/GaSb quantum wells.¹¹

Previous works derived a power-law behavior of the thickness-dependent conductivity, namely, $\sigma \propto d^s$.^{14,19} For metallic films $s=2.1$ – 2.3 ¹⁹ as long as $\xi q_F \ll 1$ with ξ the correlation length and q_F the Fermi wave vector, while for semiconducting films $s=6$.⁸ The form of the roughness correlation function plays a significant role when $\xi q_F > 1$.¹⁴ Moreover, for self-affine surfaces the roughness exponent H ($0 < H < 1$) that describes the degree of interface irregularity at short length scales ($< \xi$) has a significant influence on electron conduction.¹⁸ In any case, for a film with a smooth surface, increment of the film thickness leads to higher film conductivity. However, the development of boundary roughness with increasing thickness reduces the conductivity due to boundary scattering. Therefore, the conductivity evolution as a function of film thickness is strongly dependent on the competition between the effects due to thickness increment and roughness variation (neglecting scattering by impurities, defects, and grain boundaries). Furthermore, different film preparation conditions yield a wide variety of surface morphologies, which are inherently related to the film growth mechanisms.²¹

Indeed, noise induced roughening can lead to the formation of a self-affine rough morphology,²¹ which can evolve with increasing film thickness. For this type of growth the roughness amplitude evolves as $w \propto d^\beta$ with β the growth exponent, and the lateral correlation length as $\xi \propto d^{1/z}$ with z as the dynamic exponent.²¹ For normal self-affine growth the exponents z , H , and β satisfy the relation $z=H/\beta$. So far, scaling anomalies during dynamic growth of self-affine roughness ($z \neq H/\beta$) have not been considered in the thickness dependent conductivity for both metallic and semiconducting thin films. Indeed, former works did not consider simultaneous variation of the roughness amplitude w and the correlation length ξ with film thickness taking into account any possible relation of the corresponding scaling exponents (namely z , H , and β).¹⁸ In this paper, we explore how the dynamic growth process influences the thickness dependent conductivity in relation to anomalies (associated with violation of normal self-affine scaling, namely $z=H/\beta$) that can take place during film growth leading to faster or slower roughening with increasing film thickness.

II. Brief conductivity theory

When the bulk electron mean free path is much longer than the film thickness, proper description of the electrical conductivity requires quantum mechanics.^{14,18,20} For simplicity, we assume the free electron approximation where electrons are scattered diffusively only at rough interfaces (ignoring bulk scattering processes). The film is assumed to have thickness d with a rough film/vacuum interface at $z_2(\mathbf{r})=d/2+h(\mathbf{r})$, while the flat film/substrate interface is at $z_1=-d/2$. $h(\mathbf{r})$ are the random roughness fluctuations, which are assumed to be single-valued functions of the in-plane position vector $\mathbf{r}=(x,y)$. Moreover, we assume an isotropic auto-correlation function $C(\mathbf{r})=\langle h(\mathbf{r})h(0) \rangle$ ($\langle h(\mathbf{r}) \rangle=0$). In the Born approximation the in-plane conductivity is given by^{14,20}

$$\sigma = \frac{4e^2}{\hbar d} \sum_{\nu=1}^N \sum_{\nu'=1}^N (E_F - \varepsilon_\nu)(E_F - \varepsilon_{\nu'}) [C(E_F)_{\nu\nu'}]^{-1}, \quad (1)$$

where the matrix elements $[C(E_F)]_{\nu\nu'}$ are given by

$$[C(E_F)]_{\nu\nu'} = \delta_{\nu\nu'} \sum_{\mu=1}^N q_\nu^2 L^{\nu\mu} \int_0^{2\pi} \langle |h(q_{\nu\mu})|^2 \rangle d\theta - q_\nu q_{\nu'} L^{\nu\nu'} \int_0^{2\pi} \langle |h(q_{\nu\nu'})|^2 \rangle \cos \theta d\theta. \quad (2)$$

with $L^{\nu\mu} = U_2^2 \Psi_\nu^2(z_2) \Psi_\mu^2(z_2) (z_2 = d/2)$. N is the number of occupied minibands and E_F is the Fermi energy. $\langle |h(q)|^2 \rangle$ is the Fourier transform of the autocorrelation function $C(r)$. $q_{\nu\nu'} = (q_\nu^2 + q_{\nu'}^2 - 2q_\nu q_{\nu'} \cos \theta)^{1/2}$ with $q_\nu = [(2m/\hbar^2)(E_F - E_\nu)]^{1/2}$ being the wave vector of the ν th miniband edge, and θ is the angle between $\mathbf{q}_\nu$ and $\mathbf{q}_{\nu'}$. $\Psi_\nu(z)$ is the wave function in the z direction for smooth boundaries, and U_2 is the confining potential at the rough interface.¹⁴ The validity of the present formalism requires the roughness amplitude $w = \sqrt{\langle h^2 \rangle}$ to be much smaller than the film thickness d ($w \ll d$).^{8,14} The parameters N and E_F for a charge density n and film thickness d are given by¹⁸ $nd = (m/\pi\hbar^2)(NE_F - \sum_{\nu=1,N} E_\nu)(E_\nu < E_F)$ assuming a two-dimensional free electron gas. For electrons localized in a film with an infinite confining potential ($U_{1,2} \rightarrow +\infty$) $E_\nu = (\hbar^2/2m)(\nu\pi/d)^2$ and $L^{\nu\mu} = (\hbar^2/4md^3)^2 \nu^2 \mu^2$.^{18,20}

III. Results Discussion

As Eqs. (1) and (2) indicate the calculation of the electrical conductivity requires knowledge of $\langle |h(q)|^2 \rangle$. For self-affine fractals the roughness spectrum $\langle |h(q)|^2 \rangle$ is characterized by the power law scaling behavior, namely, $\langle |h(q)|^2 \rangle \propto q^{-2-2H}$ if $q\xi \gg 1$ and $\langle |h(q)|^2 \rangle \propto \text{const } q\xi \ll 1$.²¹ This scaling behavior is satisfied by the Lorentzian model $\langle |h(q)|^2 \rangle = (2\pi w^2 \xi^2)/(1 + aq^2 \xi^2)^{1+H/2}$ with $a = (1/2H) \times [1 - (1 + aQ_c^2 \xi^2)^{-H}]$ if $0 < H < 1$, and $a = (1/2)\ln(1 + aQ_c^2 \xi^2)$ if $H = 0$. For other correlation models see Ref. 23. $Q_c = \pi/a_0$ with a_0 a lower length scale cutoff of the order of atomic dimensions. Note that small values of H (~ 0) characterize extremely jagged or irregular surfaces, while large values H (~ 1) surfaces with smooth hills and valleys.^{21,24}

For normal self-affine growth which takes place for constant roughness exponents H (independent of film thickness) and w and ξ evolving as power laws $w \propto d^\beta$ and $\xi \propto d^{1/z}$ with $z = H/\beta$,²¹ the local surface slope $\rho = \sqrt{\langle |\nabla h|^2 \rangle}$ remains thickness invariant of the growing front (assuming constant deposition rate) because it scales as $\rho \propto w/\xi^H$.²⁰ Indeed, upon substitution of the model for $\langle |h(q)|^2 \rangle$, the local slope ρ is given by²⁴

$$\rho = \left\{ \int_{0 < q < Q_c} q^2 \langle |h(q)|^2 \rangle \frac{d^2 q}{(2\pi)^2} \right\}^{1/2} = \frac{w}{\xi} \frac{1}{\sqrt{2a}} \left\{ \frac{1}{1-H} [(1 + aQ_c^2 \xi^2)^{1-H} - 1] - 2a \right\}^{1/2}. \quad (3)$$

If, however, we assume that the correlation length evolves,

FIG. 1. Metallic films: Conductivity σ vs thickness d for specified roughness exponents (a) $H=0.3$ and (b) $H=0.9$ and values of $c=0.5, 1, 2, 3, 4, 5$ leading to consecutive conductivity decrement as can be seen clearly for large thickness d .

e.g., with a different power law, say $\xi \propto d^{1/z}$ with $z = cH/\beta$ ($z \neq H/\beta$) the corresponding local slope will evolve with film thickness as $\rho \propto d^{\beta(1-1/c)}$ leading to fast surface roughening if $c > 1$ and smoothing if $c < 1$.

The conductivity calculations were performed with $a_0 = 0.3$ nm, electron volume density $n = n_{\text{met}} = 4.8 \times 10^{-1} \text{ nm}^{-3}$ for metallic films so that many minibands to be occupied, and for semiconducting films an areal density $n_s (=nd) = n_{s(\text{sem})} = 4.8 \text{ nm}^{-2}$ so that only one miniband to be occupied. For the film surface we assumed the power laws $w = 0.05d^\beta$ (nm) with $\beta = 0.25$ ($\beta < 1$ so that $w/d \ll 1$ to ensure applicability of the formalism), and $\xi = 1.0d^{1/z}$ (nm) with $z = cH/\beta$.

A. Metallic films

Since the electron density for metallic films is high, the number of occupied minibands N can be very large. The conductivity is determined from both the intraminiband and interminiband scattering processes. For intraminiband scattering the frequency region is from 0 to $2q_\nu$, while for the interminiband scattering it is from $q_m - q_\nu$ to $q_m + q_\nu$ ($m, \nu \leq N$). The conductivity as a function of film thickness d shows pronounced QSE oscillations in the small thickness range (Fig. 1). Generally, the QSE oscillations appear when a miniband edge E_ν crosses the Fermi level E_F opening a scattering channel that leads to conductivity drop. The oscillation period is half the Fermi wavelength ($\lambda_F/2$).^{18,20}

Figure 1 shows the behavior of the conductivity for various values of the parameter “ c ” ($z = cH/\beta$). For large roughness exponents ($H \sim 1$) increment of c leads to surface roughening since the local slope increases with film thick-

FIG. 2. Metallic films: Conductivity σ vs thickness d for two distinct values of the factor $c=2, 10$ and various roughness exponents H : (a) $H=0.1$ solid line, $H=0.2$ dashes, $H=0.3$ dots, $H=0.6$ line-square, and $H=0.9$ line-cross and (b) $H=0.1$ solid line, $H=0.3$ dashes, $H=0.6$ dots, and $H=0.9$ line-cross.

ness as $\rho \propto d^{\beta(1-1/c)}$ (for $c > 1$) therefore yielding lower conductivity. This is in agreement with the fact that rougher surfaces lead to stronger scattering of the charge carriers and therefore to lower conductivity. However, comparison of Figs. 1(a) and 1(b) shows that the influence of changes of the parameter c becomes more prominent for smaller roughness exponents $H (< 0.5)$ or rougher surfaces at short length scales ($< \xi$) leading to higher charge scattering by boundary roughness. Also in this case the influence of the parameter c is more significant on the QSE oscillations.

Indeed, as a function of the roughness exponent H the thickness dependent conductivity shows for small roughness exponents $H (\sim 0)$ a transition from lower to higher conductivity at larger thickness as is shown in Fig. 2(a) for $H=0.1$. This behavior, which is observed for small parameters c leading to slower surface roughening (associated with a slow increment of the local slope) with film thickness, is suppressed with increasing roughness exponent H . In this case a monotonic conductivity decrement develops with decreasing H or increasing roughness at short wavelengths ($< \xi$). The influence of the roughness exponent H on the conductivity is clearly more prominent for $c \gg 1$ as Fig. 2(b) indicates. In this case we have a fast increment of the local slope at a rate determined mainly by the growth exponent β [since we have $\rho \propto d^{\beta(1-1/c)} \approx d^\beta$].

B. Semiconducting films

For semiconducting films the areal electron density $n_s (=nd)$ is low so that the number of occupied minibands is also small ($N \leq 2$), yielding minimal effects by interminiband scattering. For the areal $n_s = 4.8 \times 10^{-2} \text{ nm}^{-2}$ the criti-

FIG. 3. Semiconducting films: Conductivity σ vs thickness d for (a) roughness exponent $H=0.3$ and values of $c=0.5, 1, 3, 5$. (b) $c=2$ and various roughness exponents H as indicated.

cal thickness d_c above which the Fermi level E_F crosses the bottom of the second miniband ($N=2$) has the value $d_c = 10 \text{ nm}$. For moderate thickness $d < d_c$ only one lateral miniband is occupied ($N=1$) which will be the case here, where only wave vectors $0 < q < q_c = (8\pi n_s)^{1/2}$ contribute to the film conductivity. This is because forward scattering that contributes less to the conductivity occurs for $\theta=0$ or 2π yielding $k_{\perp}=0$ [since in this case we have $q_{11} = 2\sqrt{\pi n_s}(1 - \cos \theta)$], while backward scattering, which has the largest contribution to the conductivity, occurs for $\theta = \pi$ yielding $q_c = q_{11} = 2\sqrt{2\pi n_s}$.¹⁸

If we compare the effects of the parameters c and H on the conductivity for semiconducting films (Fig. 3), we can infer that their influence is significant as in the case of metallic films. However, in Fig. 3(b) we observe that the conductivity increases with decreasing roughness exponent H , which is opposite to what would be expected for rougher surfaces (also at short length scales or $< \xi$). However, for semiconducting films it has been shown that the conductivity decreases with increasing roughness exponent H (assuming fixed film thickness d)¹⁸ for small correlation lengths, so that $\xi < \lambda_F/4$.¹⁸ In addition, with increasing roughness exponent H the roughness amplitude w increases faster than the correlation length ξ (since $\xi \propto d^{\beta/cH}$), leading to higher scattering and thus to lower conductivity.

Therefore in the case of semiconducting films the evolution of the long wavelength roughness parameters w and ξ can overcome any smoothing effect due to increment of the roughness exponent H at short wavelengths yielding a decreasing conductivity with increasing thickness. This occurs when the dynamic evolution leads to rapid roughening, as for example in the case of dynamic evolution with exponents $z = cH/\beta$ and $c > 1$. The inverse behavior as a function of the roughness exponent H for metallic films indicates dominance

of short wavelength roughness ($<\xi$) due to interminiband scattering. This is not the case for semiconducting films where only one miniband is occupied, indicating absence of interminiband scattering. Nonetheless we should note that this behavior can take place up to some thickness d_c , after which more minibands are starting to be occupied, leading effectively to stronger interminiband scattering. At any rate, dynamic roughness evolution and interminiband scattering can interact and strongly influence the thickness evolution of the film conductivity.

We should point out that the conductivity model includes the following simplifications. The confining potential is assumed infinite on both sides of the structure, and it is not taken into account electron scattering on impurities and grain boundaries. In a general case the situation is more complex, and these factors should also be considered. Further, in some cases the boundary conditions on both sides of the film are significantly different and this asymmetry should also be taken into account. The influence of the confining potential on the electrical conductivity of single semiconducting films was already studied by Gottinger *et al.*,²⁵ where it was shown that the weaker the confining potential the smaller is the surface contribution to the resistivity.

IV. Conclusions

In summary, it has been shown that the dynamic evolution of surface/interface roughness during the growth process can

strongly influence electrical properties of thin films. For metallic films if the roughness amplitude w increases faster than the correlation length ξ , which is the case with increasing roughness exponent H , leading to higher charge scattering rates and thus lower conductivity, the latter still increases with increasing roughness exponent H . This is due to the presence of interminiband scattering, which enforces dominance of short wavelength roughness ($<\xi$). The opposite behavior was observed for semiconducting films where only one miniband is occupied, excluding any effects from interminiband scattering. Therefore, dynamic roughness evolution and interminiband scattering can interact and influence the film conductivity. For semiconducting films the evolution of the long wavelength roughness parameters w and ξ can overcome the smoothing effect due to increment of the roughness exponent H at short wave lengths, and decrease the thickness dependent conductivity in the case of rapid roughening (e.g., when $z=cH/\beta$ with $c>1$).

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