

## Surface roughness and grain boundary scattering effects on the electrical conductivity of thin films

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In this work, we investigate surface/interface roughness and grain boundary scattering effects on the electrical conductivity of polycrystalline thin films in the Born approximation. We assume for simplicity a random Gaussian roughness convoluted with a domain size distribution  $\sim e^{-\pi r^2/\zeta^2}$  to account for finite grain size effects with  $\zeta$ , the average domain size. For semiconducting quantum wells a peculiar interplay takes place between quantum mechanical and roughness-grain boundary scattering effects as a function of the domain size  $\zeta$  and the roughness correlation length  $\xi$ . For metallic films grain boundary scattering becomes significant for domain sizes comparable to the roughness correlation length  $\xi$ . [S0163-1829(98)05236-9]

Experiments have already manifested the substantial influence of surface/interface roughness on the conductivity of thin metallic films.<sup>1</sup> The problem was first encountered by Thompson,<sup>2</sup> and later by Fuchs<sup>3</sup> who formulated the first quasiclassical theory of the size effects. Further development of the Fuchs theory resulted in more realistic integral-type boundary conditions.<sup>4</sup> However, recent transport experiments in metallic<sup>5,6</sup> and semiconducting films<sup>7</sup> gave evidence for new features that required quantum-mechanical interpretation. The first quantum-mechanical derivations<sup>8</sup> were applied to semiconducting films.<sup>7</sup> Later, they were followed by Green functions formulations,<sup>9</sup> and coupled Boltzmann-like equations.<sup>10-12</sup>

The authors of Ref. 11 explained the universal conductivity ( $\sigma$ ) power law  $\sigma \propto d^c$  ( $c \approx 2.3$ ), which was observed in metal-like ultrathin films (<20 nm in thickness).<sup>5</sup> A similar law with  $c \approx 6$  was also found for semiconducting quantum wells.<sup>6,7,11</sup> The same authors showed that the form of the roughness correlation function plays a significant role in the limit of  $k_F \xi \gg 1$  (Ref. 12) with  $k_F$  the Fermi wave vector and  $\xi$  the roughness in-plane correlation length. In this limit the mean variation of  $s$  with film thickness  $d$  cannot be approximated by a power law. Recently, surface/interface roughness fractality effects on thin-film conductivity were studied extensively for any degree of roughness irregularity<sup>13</sup> (where finite confining potential and bulk scattering effects were also taken into account<sup>14,15</sup>) and were shown to influence significantly electron roughness scattering.<sup>13</sup>

So far the model calculations of the conductivity considered to a great extent the case of electron scattering by surface/interface roughness. An additional important component of scattering can be that of grain boundaries in polycrystalline thin films.<sup>16</sup> Mayadas, Shatzkes, and Janak<sup>17</sup> modeled grain boundaries as a sequence of partially reflecting parallel walls separated by a random distance (grain diameter), which was taken to have a Gaussian distribution around a mean value  $\zeta$ . Although this model can be appropriate for systems where columnar growth takes place,<sup>18</sup> as observed, for example, in CoSi<sub>2</sub> films,<sup>16</sup> it does not encounter simultaneously grain-boundary and roughness effects. Therefore further investigation in this direction is required in order

to describe under a unified scheme surface/interface scattering mechanisms on transport properties of polycrystalline thin films.

This will be the topic of the present work where we will investigate grain-boundary scattering effects on the conductivity of thin films convoluted with surface/interface roughness scattering effects. Following Dutta and Sinha,<sup>19</sup> in order to account for finite grain sizes in polycrystalline films, we will consider a Gaussian distribution  $\sim e^{-\pi r^2/\zeta^2}$  (with  $\zeta$  the average domain size) (Refs. 19 and 20) of domains aligned parallel to each other, and with the domain terrace to be rough. The domain roughness will be described for simplicity as Gaussian roughness.<sup>21</sup> It is anticipated that as the average grain size  $\zeta$  becomes larger than the roughness correlation length  $\xi$ , the contributions to the conductivity due to grain boundary scattering will attenuate to a degree that will be investigated in the following sections. Nevertheless, in our modeling we ignore orientational contributions from the grain-boundary shape (e.g., tilt and twist effects), and we mainly concentrate on the finite grain size effect, which limits effectively our calculations to low-angle grain boundaries.

The bottom and top boundaries of the conducting film are defined by the equations  $z = -d/2$ , and  $z = d/2 + h(\mathbf{r})$ , respectively, where we assume for simplicity reasons (as in Ref. 11) only the upper boundary rough with the roughness described by the single-valued random function  $h(\mathbf{r})$  of the in-plane position vector  $\mathbf{r} = (x, y)$ . Moreover, the roughness is assumed isotropic such that the height-height correlation function  $C(r) = \langle h(\mathbf{r}')h(\mathbf{r}'') \rangle$  depends only on the relative distance  $r = |\mathbf{r}' - \mathbf{r}''|$ . Under the assumption that only surface morphology contributes to the electron scattering, the film conductivity is given in the Born approximation by the expression<sup>11</sup>

$$\sigma(d) = \frac{e^2 \hbar^3}{m^2 d} \sum_{v=1}^N \sum_{v'=1}^N k_v^2 k_{v'}^2 [C^{-1}]_{vv'}, \quad (1)$$

$$C_{vv'} = \delta_{vv'} A_v k_v^2 \left[ \sum_{m=1}^N A_m (F_1)_{mv} \right] - A_v A_{v'} k_v k_{v'} (F_2)_{vv'}, \quad (2)$$

$$(F_1)_{mv} = \int_0^{2\pi} \langle |h(k_{mv})|^2 \rangle d\theta,$$

$$(F_2)_{vv'} = \int_0^{2\pi} \langle |h(k_{vv'})|^2 \rangle \cos \theta d\theta \quad (3)$$

with  $k_{vv'} = (k_v^2 + k_{v'}^2 - 2k_v k_{v'} \cos \theta)^{1/2}$ . The matrix elements  $C_{vv'}$  in Eq. (2) are determined by the interminiband and intraminiband transitions due to electron scattering.  $N$  is the number of occupied minibands, and  $k_v = [(2m/\hbar^2)(E_F - E_v)]^{1/2}$  with  $E_F$  and  $E_v$  being respectively the Fermi energy and the energy minimum of the  $v$ -miniband edge. If the electrons are localized by an infinite confining potential well,<sup>11</sup>  $A_v = \hbar^2 \pi^2 v^2 / md^3$  and  $E_v = (\hbar^2 / 2m)(v \pi / d)^2$ . For a film of given thickness  $d$  and carrier density  $n$ ,  $E_F$  and  $N$  are determined by the condition  $nd = (m/\pi \hbar^2)(NE_F - \sum_{v=1,N} E_v)$ .<sup>13,15</sup> Finally,  $\langle |h(k)|^2 \rangle$  is the Fourier transform of the height-height correlation function  $C(r)$ , which is needed to evaluate further the film conductivity limited by surface/interface scattering mechanisms.

A random Gaussian rough surface is characterized by the rms roughness amplitude  $\Delta$  the roughness correlation  $\xi$ , and with a roughness spectrum  $\langle |h(k)|^2 \rangle \propto \Delta^2 e^{-k^2 \xi^2 / 4}$ . The latter can be considered as a limiting case of the Fourier transform of the self-affine correlation function  $C(r) = \Delta^2 e^{-(r/\xi)^{2c}}$  (Ref. 21) for roughness exponent  $c=1$ . Furthermore, we consider the more complex surface structure of domains aligned parallel to each other, and with domain terrace to possess for simplicity Gaussian roughness in order to model polycrystalline films with finite grain size. The effect of domain sizes and shapes can be simulated through a radial Gaussian distribution function  $\propto e^{-\pi R^2 / \zeta^2}$  (Refs. 19 and 20) with  $\zeta$  the average domain size. In this case, the roughness spectrum  $\langle |h(k)|^2 \rangle_d$  is given by the simple analytic form<sup>20</sup>

$$\langle |h(k)|^2 \rangle_d = \Delta^2 \frac{\pi \xi^2 \zeta^2}{\pi \xi^2 + \zeta^2} e^{-k^2 \xi^2 \zeta^2 / 4(\pi \xi^2 + \zeta^2)}, \quad (4)$$

which incorporates finite grain size effects convoluted with grain roughness in a simple manner. Since  $\langle |h(k)|^2 \rangle_d \sim \Delta^2$ , the conductivity has the trivial dependence on  $\Delta$ , namely,  $\sigma \sim \Delta^{-2}$ , while a complex dependence is expected to arise as a function of the morphological parameters  $\xi$  and  $\zeta$ . Moreover, if we define the quantities  $\Gamma_{vv'} = 1 + a \xi^2 (k_v^2 + k_{v'}^2)$ ,  $B_{vv'} = 2a \xi^2 k_v k_{v'} / \Gamma_{vv'}$ , and  $R_{vv'} = \exp[k_v k_{v'} (\xi^2 \xi^2 / \pi \xi^2 + \zeta^2)]$ , the integrals  $(F_i)_{vv'}$ , ( $i=1,2$ ) in Eq. (3) are given in this case by the analytic forms

$$(F_1)_{vv'} = \frac{2\pi^2 \Delta^2 \xi^2 \zeta^2}{\pi \xi^2 + \zeta^2} I_0 \left( \frac{k_v k_{v'} \xi^2 \zeta^2}{2(\pi \xi^2 + \zeta^2)} \right) R_{vv'},$$

$$(F_2)_{vv'} = \frac{4\pi^2 \Delta^2}{k_v k_{v'}} I_{-1} \left( \frac{k_v k_{v'} \xi^2 \zeta^2}{2(\pi \xi^2 + \zeta^2)} \right) R_{vv'}, \quad (5)$$

which in the limit  $\zeta \gg \xi$  reduce to similar expressions obtained in earlier conductivity studies.<sup>11</sup>  $I_0(x)$ ,  $I_{-1}(x)$  in Eq. (5) are the modified Bessel functions.

*One miniband occupied ( $N=1$ ):* In semiconducting quantum wells,<sup>7</sup> the areal electron density ( $nd$ ) can be rather

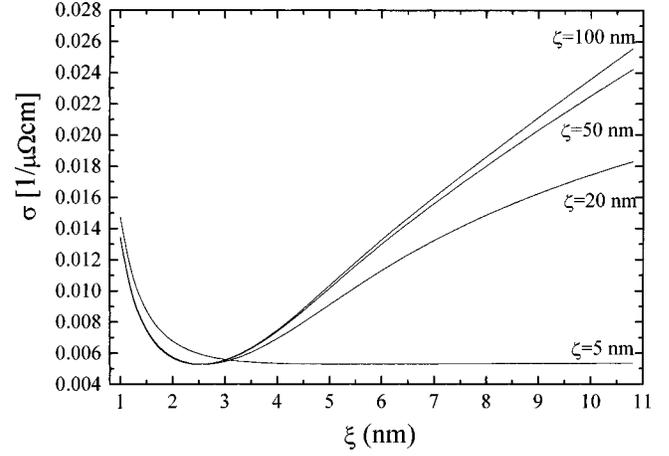


FIG. 1. Schematics of the conductivity  $\sigma$  vs  $\xi$  for Gaussian roughness for  $d=5$  nm,  $a_0=0.3$  nm,  $\Delta=0.3$  nm, and various domain sizes  $\zeta$ . Suppression of the minimum is observed for correlation lengths comparable with the grain size.

low so that the number of occupied subbands  $N$  is also small, say,  $N=1$  or 2. For  $N=1$  and assuming infinite confining potential for which  $A_v = \hbar^2 \pi^2 v^2 / md^3$ , Eqs. (1)–(5) yield the simple analytic form

$$\sigma = \frac{4nG_0}{\pi^2 \Delta^2 \xi^2} e^{k^2 \xi \zeta^2 / 2(\pi \xi^2 + \zeta^2)} \left[ \frac{2\pi^2 \zeta^2}{\pi \xi^2 + \zeta^2} I_0 \left( \frac{k^2 \xi^2 \zeta^2}{2(\pi \xi^2 + \zeta^2)} \right) - \frac{4\pi^2}{(k\xi)^2} I_{-1} \left( \frac{k^2 \xi^2 \zeta^2}{2(\pi \xi^2 + \zeta^2)} \right) \right]^{-1} d^6, \quad (6)$$

with  $G_0 = e^2 / 2\pi \hbar$  and  $k = [4\pi nd]^{1/2}$ . From Eq. (6) we obtain the power-law behavior  $\sigma \propto d^6$ , which has been observed in semiconducting quantum wells.<sup>7</sup> The numerical calculations were performed for carrier density  $n = 4 \times 10^{-2}$  nm<sup>-2</sup>, rms roughness amplitude  $\Delta = 0.3$  nm, and film thicknesses lower than 10 nm in order that only one miniband be occupied ( $N=1$ ).<sup>11</sup>

Figure 1 shows the conductivity  $\sigma$  versus  $\xi$  for Gaussian roughness and various average domain sizes  $\zeta$ . A minimum is observed at a correlation length  $\xi \sim \lambda_F / 4$  (with  $\lambda_F \sim d$  the Fermi wavelength), which indicates selectively strong scattering at a particular value of  $\xi$ .<sup>7,13</sup> This is due to the fact that  $\langle |h(k)|^2 \rangle_d$  increases as  $\sim \xi^2$ , reaches a maximum, and finally decreases with further increment of  $\xi$  as  $\sim e^{-\xi^2}$ . At large correlation lengths  $\xi \sim \lambda_F$  (for constant amplitude  $\Delta$ ), a normal behavior is achieved, which corresponds to weaker surface electron scattering with decreasing ratio  $\Delta/\xi$  (surface smoothing).<sup>13</sup> However, the minimum occurs if the average domain size  $\zeta$  is significantly larger than the roughness correlation length  $\xi$ . For small domain sizes the minimum becomes weaker and broader, as well as cease to exist for average domain sizes  $\zeta \leq \lambda_F$  leading to dominance of grain finite size scattering effects.

Figure 2 shows  $\sigma$  versus  $\zeta$  for fixed correlation length  $\xi$ . The dependence of the conductivity on the average domain size  $\zeta$  becomes rather complex for small correlation lengths  $\xi (< \lambda_F / 2)$  where a continuous decrement of  $\sigma$  with  $\zeta$  is observed. As the correlation length increases a minimum appears, which is followed by a rather slow increment of the

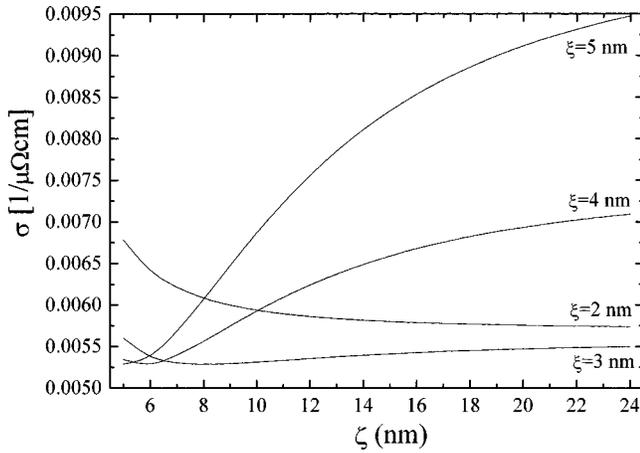


FIG. 2. Schematics of the conductivity  $\sigma$  vs  $\zeta$  for Gaussian roughness for  $d=5$  nm,  $a_0=0.3$  nm,  $\Delta=0.3$  nm, and various correlation lengths  $\xi$  as indicated. A complex dependence of the conductivity develops for small correlation lengths ( $\xi \sim \lambda_F/2$ ;  $\lambda_F \sim d$ ).

conductivity as long as  $\xi < \lambda_F$ . Finally, for large correlation lengths  $\xi > \lambda_F$ , a rapid increment of the conductivity is observed as a function of  $\zeta$ , which is accompanied with the disappearance of the minimum at small  $\zeta$ . Therefore, the conductivity of semiconducting quantum wells is altered with the presence of finite-size grain boundary scattering convoluted with roughness electron scattering for small grain sizes.

*Metallic films* ( $N \gg 1$ ): Our calculations were performed for carrier density  $n \approx 3 \times 10^{17} \text{ nm}^{-3}$ , which is typical for  $\text{CoSi}_2$ , rms roughness amplitude  $\Delta=0.3$  nm, and domain sizes  $\zeta > 3$  nm, which have been observed in metal-like polycrystalline  $\text{CoSi}_2$  films.<sup>16,18</sup> The influence of the roughness parameters  $\Delta$  and  $\xi$  on the conductivity was shown already in earlier studies.<sup>10–13</sup> We remark that smoothing of the surface/interface ( $\Delta/\xi \ll 1$ ) leads to weak electron scattering by roughness irregularities that can enhance drastically the film conductivity even by more than an order of magnitude.

In Fig. 3 we present plots of the conductivity  $\sigma$  versus  $\xi$  for various domain sizes  $\zeta$ . The conductivity is increasing

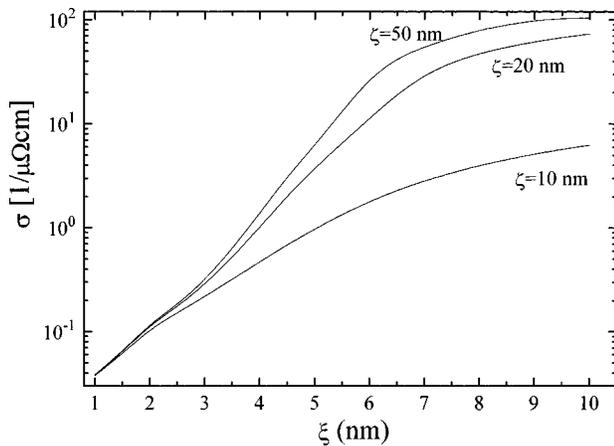


FIG. 3. Schematics of film conductivity  $\sigma$  vs  $\xi$  for Gaussian roughness,  $d=1.2$  nm,  $a_0=0.3$  nm,  $\Delta=0.3$  nm, and various domain sizes  $\zeta$  as indicated. Saturation of the conductivity and dominance of grain boundary scattering for large correlation lengths occurs ( $\xi > \zeta$ ).

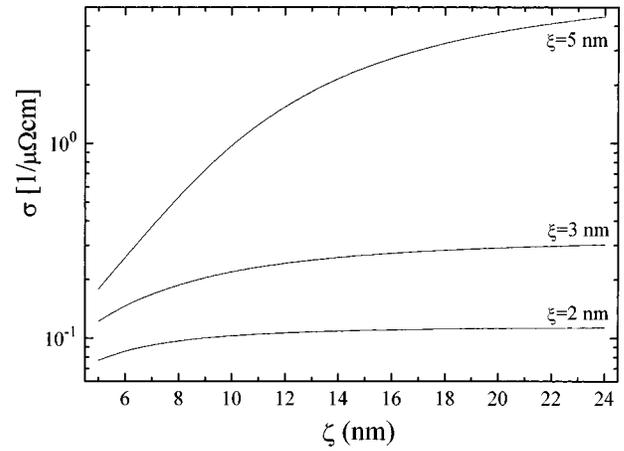


FIG. 4. Schematics of film conductivity  $\sigma$  vs  $\zeta$  for Gaussian roughness,  $d=1.2$  nm,  $a_0=0.3$  nm,  $\Delta=0.3$  nm, and various correlation lengths  $\xi$  as indicated. The saturation of the conductivity at large domain sizes occurs rapidly for small correlation lengths ( $\xi \sim \lambda_F/2$ ).

monotonically with increasing  $\xi$ , however, at a lower rate as  $\xi$  approaches values close to or larger than average domain size  $\zeta$ . Indeed, for an increment of  $\zeta$  by one order of magnitude, the conductivity can increase by more than two orders of magnitude in the regime of large correlation lengths ( $\xi > 4\lambda_F$ ). The latter indicates the strong effect that the finite-size grain boundary scattering can impose on electrical transport properties. However, saturation of the conductivity and dominance of grain boundary scattering for large correlation lengths occur rather rapidly for small domain sizes ( $\xi \gg \zeta$ ). The contribution of the domain size can be seen alternatively in Fig. 4, where we plot  $\sigma$  versus  $\zeta$  for various roughness correlation lengths  $\xi$ , where the conductivity shows a significant sensitivity for small variations of the correlation length. A rapid increase is observed for correlation lengths comparable to the film thickness, which could be larger than an order of magnitude.

In a more realistic approach finite confining potential effects have to be taken into account and bulk impurity electron scattering, as well as asymmetrical boundary conditions on both sides of the film. For semiconducting and metallic films<sup>13,22</sup> it was shown that the weaker the confining potential the smaller the surface/interface scattering contribution to the resistivity. Moreover, bulk scattering due to random impurities located in the film's interior suppresses significantly the influence of surface/interface irregularities for electron mean free paths comparable with the film thickness.<sup>13</sup> The additional effect of finite-size grain boundary scattering will limit further the influence of surface/interface roughness. Therefore, these morphological effects have to be taken carefully into account in explaining electrical transport properties in polycrystalline systems.

In our study, we correlated known information of surface/interface scattering effects on the electrical conductivity of metallic and semiconducting thin films with analytic roughness models that incorporate finite grain size and roughness effects, in order to investigate limitations imposed by finite grain size boundary scattering effects on the conductivity of polycrystalline morphologies. These scattering mechanisms cannot be separated whenever the roughness correlation

length is comparable to or larger than the grain size. In the latter case, finite-size grain boundary scattering can suppress enormously the roughness contribution on the film's conductivity to a degree that depends on the particular set of competing roughness parameters ( $\xi$  and  $\zeta$ ) that characterize the nanoscale grain morphology. Nonetheless, our calculations are limited to low angle grain boundaries ( $\Delta/\zeta \ll 1$ ), and future work should address more precisely orientational contributions of grain boundaries in order that a more complete

understanding of surface/interface electron scattering mechanisms can emerge.

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