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The roughness-induced classical size effect in thin films

R Lenk and A Knäbchen

University of Technology Chemnitz–Zwickau, Institute for Theoretical Physics, PO Box 964,
D-O9009 Chemnitz, Federal Republic of Germany

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Abstract. We consider the electronic transport process in thin films with rough surfaces. The approach employed is based on classical concepts and avoids some special assumptions used in earlier papers. Roughness is described in terms of scatterers that are distributed randomly on the surfaces. Their differential scattering cross section yields a generally angle-dependent specular parameter. A new form of this angle dependence is proposed. We show that the homogeneity of the lateral carrier distribution, as postulated by Fuchs in his well known paper published in 1938, is associated with the time-reversal symmetry of the microscopic scattering cross section. The divergence of the classical surface-dominated conductivity, i.e. for vanishing bulk scattering, is enhanced for angle-dependent specular parameters in comparison with the constant parameter introduced by Fuchs. We further show how to make contact between classical formulations and quantum-mechanical approaches leading to a stronger dependence of the conductivity on the film thickness. Our results are compared with existing classical size-effect theories.

1. Introduction

The electrical resistivity of metallic films is determined by the simultaneous occurrence of surface scattering and scattering within the film. The latter effect gives rise to a finite mean free path (MFP) l in the extended bulk. The surface contribution to the resistivity is attributed to surface roughness, strictly speaking, it is due to the deviations from a perfectly plane surface. This extra resistance gains importance as the thickness of the film d approaches the MFP, i.e., in the classical size effect regime. Theoretically, the influence of the surface on the DC conductivity in this case was first studied comprehensively by Fuchs in 1938 [1]. His calculations are based on the Boltzmann equation where the surface is incorporated via boundary conditions on the velocity distribution function. In particular, a single parameter p defines the fraction of conduction electrons reflected specularly at the surface, the remainder being scattered diffusely. This phenomenological description contains no direct information about the microscopic scattering mechanism itself. Nevertheless, Fuchs's theory is widely applied to analyse experimental data.

In the last decades, Fuchs's formulation has been subject to a number of modifications (see, for reviews, [2]–[4]). All these considerations have retained a substantial feature of the original model, namely, an additional surface scattering is superimposed on the bulk relaxation mechanism. More recently, some authors have proposed quantum mechanical approaches leading to a very pronounced dependence of the conductivity on the thickness. In the almost-classical case when a large number of lateral subbands are occupied, one finds for the surface-dominated conductivity $\sigma_s \simeq d^2$ [5–8]†, in good agreement with experimental

† We note that such a quadratic law for thin films was also found by Jaggi [9], employing, however, a distinct approach.

data for a certain thickness region. Beyond former theories we demonstrate below how to make contact between the results of a purely classical framework and this relation.

It seems to be rather surprising that a classical model starting with a detailed consideration of scattering processes at the surface has not yet been investigated. It is the aim of the present work to do this and thus to bridge, in the noted sense, the gap to quantum-mechanical results. We remark that a classical approach offers the opportunity to go beyond the perturbational treatment applied in all quantum-mechanical papers. The validity of these theories is limited (in a strict sense) to nearly specular reflecting surfaces.

In our model, the roughness is represented by scatterers that are distributed randomly over the surface. Each of them scatters particles with a differential scattering cross-section S . As a microscopic quantity this scattering cross section fulfils time-reversal symmetry, $S(\mathbf{v}', \mathbf{v}) = S(\mathbf{v}, \mathbf{v}')$, where \mathbf{v} and \mathbf{v}' refer to the velocity of an incident or emerging particle, respectively. It is this symmetry which guarantees homogeneity of the lateral carrier distribution in agreement with the classical limit of the local density of states. This property of a physically sensible solution would be violated for an asymmetric microscopic scattering law. The lateral homogeneity mentioned has been only postulated by Fuchs. Our present investigation shows, however, that this assumption conflicts with the common model of an angle-independent specularly parameter.

The normalization of the specular component is determined by the requirement of current balance, of course. As a result, our specular contribution, $p(\mathbf{v})$, exhibits in general an angular dependence and obeys the physically appealing condition $p(\mathbf{v}) \rightarrow 1$ for grazing incidence.

We mention that, apart from the symmetry as pointed out above, $S(\mathbf{v}', \mathbf{v})$ is in principle allowed to be an arbitrary angular function. This angular dependence is less important when the MFP is small compared to the thickness. Otherwise, the free propagation is scarcely restricted by internal scattering but mostly terminated by diffuse surface scattering. It is well known that the corresponding surface-dominated conductivity tends to infinity for increasing MFP. This singularity results from carriers travelling nearly parallel to the surface. Consequently, their contribution is given by the surface properties for grazing incidence. In particular, a constant specularly parameter as introduced by Fuchs yields a weak logarithmic divergence. We will show that a physically sensible angle-dependent scattering law does not remove this singularity but makes it even more pronounced.

We derive our formulae in the diffusion picture, i.e., a stationary current is related to a given carrier density gradient. This choice is only a formal matter because it is well known how to progress via the Einstein equivalence between a driving force and a negative gradient of the chemical potential from diffusivity to conductivity. Some remarks on the transcription to the familiar force case are given in the appendix.

The outline of the paper is as follows. In section 2 we compile some equations associated with the classical distribution function employed and derive the total transition probability from the scattering cross section, S . Additionally, we discuss some realizations of S . Afterwards we calculate the diffusion constant in the case when only surface scattering occurs (section 3). In section 4 the simultaneous occurrence of both surface and bulk scatterers will be considered. As expected, we rederive and confirm Fuchs's result with an angle-dependent specularly parameter. Section 5 is used for discussions of the different solutions and limiting cases. A summary is given in section 6.

2. Transition probability associated with surface roughness

Let $\varrho(\mathbf{r}, \mathbf{v})$ denote the density distribution of carriers with velocity \mathbf{v} at point \mathbf{r} and at a

fixed energy $E = \frac{1}{2}mv^2$ which implies $v = \text{constant}$. The total carrier density is then given by

$$\rho(\mathbf{r}) = \int d\Omega \rho(\mathbf{r}, \mathbf{v}) \tag{1}$$

where the integration runs over all directions of \mathbf{v} . The current density can be written as

$$\mathbf{j}(\mathbf{r}) = \int d\Omega \mathbf{v} \rho(\mathbf{r}, \mathbf{v}). \tag{2}$$

Finally, we introduce the surface scattering relation

$$\rho(\mathbf{r}_s, \mathbf{v}') = \int d\Omega f_{\pm}(\mathbf{v}', \mathbf{v}) \rho(\mathbf{r}_s, \mathbf{v}) \tag{3}$$

i.e., particles leaving the surface from a point \mathbf{r}_s with velocity \mathbf{v}' are related to those arriving with velocity \mathbf{v} via a transition probability per unit solid angle, $f_{\pm}(\mathbf{v}', \mathbf{v})$. Unlike the expressions defined above, the integral runs in equation (3) over the half sphere appropriate for the upper or lower surface. Generally, the surfaces may differ structurally corresponding to the experimental situation where the film is commonly deposited on a glass substrate in vacuum. This fact is expressed in terms of the two quantities f_+ and f_- , the transition probabilities at the upper ($z = d$) or lower ($z = 0$) surface, respectively. For the sake of brevity, however, these indices are often omitted in the following.

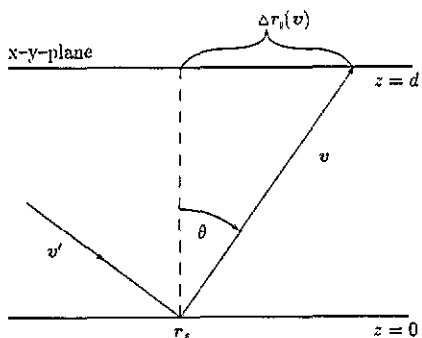


Figure 1. This picture shows the co-ordinates employed. The film is bounded by the planes $z = 0$ and $z = d$. The longitudinal position is determined by $r_{\parallel} = (x, y)$. The carrier's velocity is often expressed in terms of spherical angles (θ, φ) . For clarity, the azimuthal angle, φ , is not shown. The angle of the velocity with the surface normal, θ , varies from 0 to π . The horizontal distance between surface points where a particle emerges or arrives is denoted by $\Delta r_{\parallel}(\mathbf{v})$.

Now we ask for the transition probability characterizing a statistically rough surface. The roughness is modelled by the presence of surface scatterers, assumed to be distributed homogeneously and uncorrelated. This implies that the intensities emerging from different scatterers simply add; interference effects can be neglected. The scattered current density emerging from a single scatterer is given by (see figure 1)

$$\mathbf{j}_{\text{out}}(\mathbf{r}, \mathbf{v}') = S(\mathbf{v}', \mathbf{v}) \left[\widehat{\mathbf{r} - \mathbf{r}_s} / |\mathbf{r} - \mathbf{r}_s|^2 \right] \mathbf{j}_{\text{in}}(\mathbf{r}_s, \mathbf{v}) \tag{4}$$

where \mathbf{j}_{in} refers to particles incident with velocity \mathbf{v} and S is the differential scattering cross section. To find the total scattered current density per unit solid angle $d\Omega'$, expression (4) has to be multiplied by the mean number of scatterers belonging to the surface region

concerned, $N_s(\mathbf{r} - \mathbf{r}_s)^2 d\Omega' / |\cos \theta'|$, where N_s is their mean areal density. We are thus led to

$$(1/j_{\text{in}}) dj_{\text{out}}^{\text{total}}(\mathbf{v}')/d\Omega' = N_s S(\mathbf{v}', \mathbf{v}) / |\cos \theta'| \quad (5)$$

which is simply the transition probability associated with surface scatterers only. Taking into account the fact that incoming carriers can be also reflected specularly, the total transition probability has the form

$$f(\mathbf{v}', \mathbf{v}) = p(\mathbf{v}') \delta(\widehat{\mathbf{v}}' - \widehat{\mathbf{v}}^*) + N_s S(\mathbf{v}', \mathbf{v}) / |\cos \theta'| \quad (6)$$

where the first term includes the specular reflection of a carrier from $\mathbf{v} = (v_x, v_y, v_z)$ to $\mathbf{v}^* = (v_x, v_y, -v_z)$ and the δ function is normalized as $\int d\Omega \delta(\widehat{\mathbf{v}}' - \widehat{\mathbf{v}}^*) = 1$. The factor p , in view of Fuchs's paper [1], will be denoted as the specularly parameter. This parameter can vary from zero (for completely diffuse scattering) to unity (for completely specular reflection). Its normalization is determined by the requirement of current balance. This is related to the normal component of the current only and yields immediately (equations (2) and (3)) the relation

$$\int d\Omega' \zeta' f(\mathbf{v}', \mathbf{v}) = \zeta \quad (7)$$

where we have introduced $\zeta = |\cos \theta|$. Applying this formula to (6) one obtains

$$p(\mathbf{v}) = 1 - \frac{N_s}{|\cos \theta|} \int d\Omega' S(\mathbf{v}, \mathbf{v}') \quad (8)$$

for the generally angle-dependent specular parameter.

Provided that the transition probability is normalized according to (7), we can derive

$$\int d\Omega f(\mathbf{v}', \mathbf{v}) = 1. \quad (9)$$

This additional relation rests on the time-reversal symmetry fulfilled by the scattering cross section S . It will be used below to prove the homogeneity of the lateral carrier density distribution.

Equations (7) and (9) have already been given by Moliner and Simons [10, 11] in a somewhat different context, and applied by Greene and O'Donnell [12, 13] to consider Fuchs's theory with an angle-dependent specularly parameter. The complicated results obtained by these authors have, however, attracted little attention.

In view of its influence on the resulting diffusivity, and for its own interest, the angular dependence of the scattering cross section S should be taken seriously. To specify this dependence we assume that S is merely a function of the angles θ , θ' between \mathbf{v} , \mathbf{v}' and the normal direction of the surface, i.e., the angle of incidence or reflection, respectively. Taking into account the symmetry condition, S can now be rewritten as $S = F(\cos \theta', \cos \theta) = F(\cos \theta, \cos \theta')$. As the simplest case, we may further assume that F factorizes, cf. below. Additionally, we will consider only the simplest realizations of F , or, in more mathematical terms, refer to a power-series expansion in the variable $\cos \theta$. Obviously, a constant term is not possible. In order to compensate the denominator in equation (8), S has to include at least a linear term, i.e., $S = S_0 \cos \theta' \cos \theta$, $S_0 = \text{constant}$. This *ansatz* is not too artificial because already the effectiveness of classical surface

scattering is weighted by a cosine (Lambert's law). Consequently, that scattering law could be considered as its symmetrized representation. It yields for the specularity parameter (equation (8)) $p(v) = 1 - \pi N_s S_o$. This is the only non-trivial case leading to an angle-independent parameter p .

Nevertheless, this scattering law is not compatible with quantum mechanics where the simplest case is a $\cos \theta' \cos \theta$ behaviour of the scattering amplitude. It can be shown to be correct for a single scatterer whose size (diameter) is small compared with the wavelength [14]. For the scattered current, as a bilinear function of the scattering amplitude, we are thus led to

$$S = S_o \cos^2 \theta' \cos^2 \theta \quad S_o = \text{constant.} \quad (10)$$

We argue, guided by the arguments given above, that this representation is the simplest function that obeys all physical requirements. Equation (10) will therefore be favoured in the following. Furthermore we mention that this scattering law of a single surface scatterer was employed in [8] to confirm the result of Fishman and Calecki [7] on the quantum size effect in thin films that includes a behaviour of the conductivity as d^6 .

Substituting equation (10) into (8) one obtains

$$p(v) = 1 - \frac{2}{3} \pi N_s S_o |\cos \theta| \equiv p(\zeta). \quad (11)$$

In view of the derivation, the condition $0 \leq p \leq 1$, i.e. $2\pi N_s S_o/3 \leq 1$, is obeyed without further assumptions, since the neglect of interference effects (cf discussion following (5)) implies that the mean distance between scatterers $\sim N_s^{-1/2}$ is larger than their size $\sim S_o^{1/2}$. Since p approaches unity for grazing incidence, $\theta \rightarrow \pi/2$ (or π), particles running almost parallel to the surface are not affected by surface irregularities. These carriers will give rise to an infinite diffusivity.

3. Surface-dominated diffusion

It is obvious that, without a bulk relaxation mechanism, a particle emerging from either surface is not scattered until it arrives at the other one. This implies that for the carrier distribution

$$\varrho(\mathbf{r}, \mathbf{v}) = \text{constant along the straight path with direction } \hat{\mathbf{v}} \quad (12)$$

including the initial and final point on the corresponding surface (see, for definitions, figure 1), or, in other words, the distribution emerging from the lower surface ($v_z > 0$) yields that incident on the upper one and vice versa. The intensities incident and scattered at one and the same surface are related via the scattering relations (3).

In the current-carrying state, the distribution $\varrho(\mathbf{r}, \mathbf{v})$ is anisotropic with respect to the variable \mathbf{v} to yield a non-vanishing current density (2). Furthermore, to give the driving density gradient, $\varrho(\mathbf{r}, \mathbf{v})$ depends linearly on the longitudinal position. The (relative) shape of the lateral distribution between the planes $z = 0$ and $z = d$ remains unchanged, however. This situation is described by the *ansatz*

$$\varrho(\mathbf{r}, \mathbf{v}) = \varrho(z, \mathbf{v})(1 + g\mathbf{r}_{\parallel}) \quad g = \text{constant} \quad (13)$$

where $\varrho(z, v)$ is the distribution for a reference cross section at $r_{\parallel} = 0$. Bearing in mind formula (12), the incoming densities at both surfaces are thus related to each other via

$$\begin{aligned} \varrho_+(v_+) &\equiv \varrho(d, v_+) = [1 - g\Delta r_{\parallel}(v_+)] \int d\Omega_- f_-(v_+, v_-) \varrho_-(v_-) \\ \varrho_-(v_-) &\equiv \varrho(0, v_-) = [1 - g\Delta r_{\parallel}(v_-)] \int d\Omega_+ f_+(v_-, v_+) \varrho_+(v_+) \end{aligned} \quad (14)$$

with an 'anisotropy' term

$$g\Delta r_{\parallel}(v) = gd|\tan\theta|\cos\varphi.$$

v_{\pm} refer to $v_z \gtrless 0$. The density difference from the position where the carriers started can be found from equation (13) and is included via the angle-dependent pre-factor in brackets. These pre-factors are equal for $v_+^* = v_-$, i.e., if the variables v behave as do the velocities of a particle before and after a specular reflection. Then, provided that the scattering model (11) discussed in section 2 is employed, the system of integral equations (14) is solved by

$$\varrho_{\pm}(v_{\pm}) = [1 - g\Delta r_{\parallel}(v_{\pm})\Phi_{\pm}(\zeta_{\pm})]\bar{\varrho}. \quad (15)$$

These distributions are characterized by an anisotropy corresponding to the current-carrying state. For $g = 0$, this formula has to be compatible with the current-free result and thus $\bar{\varrho}$ is the equilibrium distribution at the surfaces. Employing formula (9), equations (14) are solved in this case by $\bar{\varrho} = \text{constant}$. Furthermore, without a carrier density gradient, the density distribution is of course constant with respect to the longitudinal position $r_{\parallel} = (x, y)$. Finally, if we take advantage of the fact that the carriers cannot be scattered until they arrive at one of the surfaces (equation (12)) we find immediately

$$\varrho(r, v) = \text{constant} \equiv 1 \quad \text{for } g = 0 \quad (16)$$

i.e., the equilibrium density distribution is laterally homogeneous. As an implication of relation (9), this solution rests on the symmetry condition fulfilled by the differential scattering cross section S . We note that result (16) remains valid even for surfaces with different specularly parameters p_+ and p_- . To avoid confusion, we emphasize again that (9) is an additional relation following from the symmetry of S and is not identical with the normalization condition (7).

For $g \neq 0$, the contribution of the anisotropy term in (14) to the diffusely scattered part vanishes since $\int d\Omega \cos\varphi = 0$. There remains a set of linear equations that yield the still unknown $\Phi_{\pm}(\zeta)$ as functions of the specularly parameters $p_{\pm}(\zeta)$. To linear order in the density gradient $\sim g$, we obtain

$$\Phi_{\pm}(\zeta) = [1 + p_{\mp}(\zeta)]/[1 - p_{-}(\zeta)p_{+}(\zeta)] \quad (17)$$

thus the density distributions (15) incident upon either surface are now determined.

Using ϱ_{\pm} the density distribution within the film can be constructed directly. In particular we find that

$$\begin{aligned} \varrho(z, v_+) &= \{1 + [(d-z)/d]g\Delta r_{\parallel}(v_+)\}\varrho_+(v_+) \\ &= 1 + [(d-z/d - \Phi_+(\zeta_+)]g\Delta r_{\parallel}(v_+) \\ \varrho(z, v_-) &= [1 + (z/d)g\Delta r_{\parallel}(v_-)]\varrho_-(v_-) = 1 + [z/d - \Phi_-(\zeta_-)]g\Delta r_{\parallel}(v_-). \end{aligned} \quad (18)$$

These solutions interpolate simply between the boundary distributions. Obviously, the anisotropy terms do not contribute to the angularly averaged density and thus the total density (1) of the reference cross section is given by $\varrho(z, \mathbf{r}_\parallel = \mathbf{0}) = 4\pi$ and $\mathbf{g} = (4\pi)^{-1} \partial\varrho(\mathbf{r})/\partial\mathbf{r} \equiv (4\pi)^{-1} \text{grad}\varrho(\mathbf{r})$. The resulting current density (2) becomes independent of z , i.e., surface scattering alone leads to a laterally homogeneous current distribution. The corresponding diffusion constant,

$$D_s = \frac{dv}{4} \int_0^1 d\xi \frac{1 - \xi^2}{\xi} \left(\frac{[1 + p_-(\xi)][1 + p_+(\xi)]}{1 - p_-(\xi)p_+(\xi)} \right) \tag{19}$$

is defined by Fick's law, $\mathbf{j} = -D \text{grad}\varrho$. This result for surface-dominated diffusion will be discussed further in section 5.

4. Diffusion against surface and bulk scatterers

The general case is characterized by the simultaneous occurrence of both surface and bulk scatterers. The latter are assumed to be distributed homogeneously and to scatter isotropically. Their mean volume density and their scattering cross section give rise to a finite mean free path, l .

Equation (12) is now invalid: $\varrho(\mathbf{r}, \mathbf{v})$ changes along any straight line in direction $\hat{\mathbf{v}}$, i.e., with the co-ordinate $s = \hat{\mathbf{v}}\mathbf{r}$, according to

$$(\partial/\partial s + 1/l)\varrho(\mathbf{r}, \mathbf{v}) = (1/4\pi l)\varrho(\mathbf{r}). \tag{20}$$

The left-hand term accounts for scattering out of the ray direction, the right-hand one for scattering in that direction from all others with equal weight. The solution of (20) with the corresponding boundary values reads

$$\begin{aligned} \varrho(\mathbf{r}, \mathbf{v}_+) &= e^{-(z/\zeta_+ l)} \varrho_-(\mathbf{r}_s^+, \mathbf{v}_+) + \frac{1}{4\pi} \int_0^z \frac{dz'}{\zeta_+ l} e^{[-(z-z')/\zeta_+ l]} \varrho(\mathbf{r}_+(z-z')) \\ \varrho(\mathbf{r}, \mathbf{v}_-) &= e^{-(d-z)/\zeta_- l} \varrho_+(\mathbf{r}_s^-, \mathbf{v}_-) + \frac{1}{4\pi} \int_z^d \frac{dz'}{\zeta_- l} e^{[-(z'-z)/\zeta_- l]} \varrho(\mathbf{r}_-(z'-z)) \end{aligned} \tag{21}$$

with

$$\mathbf{r}_\pm(t) = \mathbf{r} - (t/\zeta)_\pm \hat{\mathbf{v}}_\pm \quad \mathbf{r}_s^+ = \mathbf{r}_+(z) \quad \mathbf{r}_s^- = \mathbf{r}_-(d-z).$$

The integrations in (21) are to be performed along a ray from the point $\mathbf{r} = (\mathbf{r}_\parallel, z)$ in the direction $-\hat{\mathbf{v}}_\pm$ to the lower or upper surface, respectively. As before in section 3, the boundary values of the outgoing intensities, $\varrho_\pm(\mathbf{r}_s, \mathbf{v}_\mp)$, are determined by the corresponding surface scattering relation. In the equilibrium case, $\varrho(\mathbf{r}, \mathbf{v}) \rightarrow \varrho(z, \mathbf{v})$, the solution is again found to be $\varrho(\mathbf{r}, \mathbf{v}) \equiv \text{constant} \equiv 1$. Generally, i.e., with a longitudinal density gradient, we can determine firstly the incident intensities at both boundaries. The total density $\varrho(\mathbf{r})$ remains unaffected by the anisotropy of the velocity distribution. The result, for the special case of equivalent surfaces, $p_+(\zeta) = p_-(\zeta) \equiv p(\zeta)$, is

$$\varrho(\mathbf{r}_s, \mathbf{v}_\pm) = 1 - \left\{ \zeta_\pm l [1 - e^{(-d/\zeta_\pm l)}] / d [1 - p(\zeta_\pm) e^{(-d/\zeta_\pm l)}] \right\} \mathbf{g} \Delta \mathbf{r}_\parallel(\mathbf{v}_\pm). \tag{22}$$

The limiting cases can be easily extracted: For $d \ll \zeta l$, equations (18) are simply reproduced; for $d \gg l$, the film thickness d in (18) has to be replaced by ζl .

The boundary values (22) now given, the basic equations (21) determine straightforwardly the distribution everywhere as

$$q(z, v_+) = 1 - (\zeta_+ l / d) \left[1 - \left\{ 1 - [p(\zeta_+)] / [1 - p(\zeta_+) e^{(-d/\zeta_+ l)}] \right\} e^{(-z/\zeta_+ l)} \right] g \Delta r_{\parallel}(v_+) \quad (23)$$

and an analogous expression for $q(z, v_-)$ with z replaced by $d - z$. Remember that z in (23) is the distance from the lower surface. The first term 1 in the brackets describes the pure bulk effect.

The current density is laterally *not* constant. According to (23), the contribution of particles with fixed ζ comprises a roughness-induced term $\sim -\{e^{(-z/\zeta l)} + e^{[-(d-z)/\zeta l]}\}$. For a thick film, this means a reduced current in a surface layer of thickness l . For a thin film, this reduction spreads over the whole cross section, leaving a constant current density again for pure surface scattering as in section 3.

If we define a z -dependent diffusivity via $j(z) = -D(z) \text{grad } q$ we find that

$$D(z) = D_b - \frac{vl}{4} \int_0^1 d\zeta \zeta (1 - \zeta^2) \frac{1 - p(\zeta)}{1 - p(\zeta) e^{(-d/l\zeta)}} \{e^{(-z/l\zeta)} + e^{[-(d-z)/l\zeta]}\} \quad (24)$$

where $D_b = vl/3$ is the diffusivity of a 3D bulk. The mean diffusion constant, $D \equiv (1/d) \int_0^d dz D(z)$, reads

$$\frac{D}{D_b} = 1 - \frac{3}{2\alpha} \int_0^1 d\zeta \zeta (1 - \zeta^2) \frac{[1 - p(\zeta)] [1 - e^{(-\alpha/\zeta)}]}{1 - p(\zeta) e^{(-\alpha/\zeta)}} \quad \alpha = \frac{d}{l} \quad (25)$$

which is simply the well known Fuchs-Sondheimer formula [1, 15] with an angle-dependent specularly parameter. Substituting the special parameter (11), this expression is approximated in the limit of thick films by

$$D/D_b \approx 1 - (2\pi l/15d) N_s S_0 \quad l/d \ll 1 \quad (26)$$

i.e., only a small correction from surface roughness remains. Similarly, for the Fuchs model of a constant parameter p_F , one obtains

$$D/D_b \approx 1 - (3l/8d) (1 - p_F). \quad (27)$$

Thus, for $l \ll d$, one can identify the quantity $\frac{16}{45} N_s S_0$ with the fraction $1 - p_F$ of diffusely scattered carriers in the original theory.

5. Discussion

The calculations presented in the previous sections are based on the diffusion picture. For completeness, some remarks on the transition to the more familiar force case are summarized

in the appendix. For a sharp Fermi energy, D/D_b can be replaced by σ/σ_b with σ as the common conductivity.

Let us at first consider expression (19). It is obvious that, independent of the detailed nature of the specular parameter, the surface-dominated diffusion constant shows a grazing incidence divergence for $\zeta \rightarrow 0$. This well known result can be understood by keeping in mind the fact that classical particles are allowed to fly parallel or nearly parallel to the surfaces of the film. This implies that a classical surface resistance does not arise unless a bulk relaxation mechanism exists.

Provided that the specular parameter has no angular dependence, $p = \text{constant}$, one obtains from (19) a weak logarithmic singularity. Such a singularity was already found by Fuchs in his 1938 paper [1]. Generally, the diffusion constant shows a stronger divergence for specular parameters which approach unity for grazing incidence. In particular, when p is given by (11), D diverges as ζ^{-1} . Therefore the grazing incidence singularity does not result from a crude description of surface scattering, but is even more pronounced for more realistic models.

As opposed to a classical framework, quantum mechanical carrier propagation in a thin film is mainly characterized by the underlying discrete energy-level spectrum set up by the lateral boundaries [6, 16]. The discreteness of the eigenvalues becomes more and more negligible as the number of occupied subbands increases. There remains, however, an important difference from the classical case, namely, there is always a non-vanishing lateral component of the wavevector, $k_z > 0$. Further, we can associate the ratio of this component to the total wavevector, k , with an classical angle of incidence according to $\theta = \cos^{-1}(k_z/k)$. The minimal value, $k_z = \pi/d$, determines an upper limit, smaller than $\theta = \pi/2$ in equation (19), for the angle of incidence and thus prevents a divergence of the surface-dominated conductivity. In particular, substituting the limit of integration into (19) yields (to leading order in $kd \gg 1$)

$$\sigma_s \simeq d^2/N_s S_0. \quad (28)$$

We emphasize that the angular dependence of $p(\zeta)$, equation (11), is essential for this result. This quadratic law already mentioned in the introduction is common to a number of quantum mechanical investigations [5–8].

As a result derived from perturbational methods the resistivity is linearly dependent on the roughness parameter, $\sigma_s^{-1} \simeq N_s S_0$. A comparison with the general expression (19) shows, however, that a classical treatment has the advantage of going beyond this approximation and describing also the non-linear dependence on $N_s S_0$.

The introduction of an upper limit $\theta < \pi/2$ for the angle of incidence as discussed above is of relevance unless the free propagation of the carriers is restricted to finite free paths due to the very existence of bulk scattering. The conductivity in this case is given by formula (25). To obtain an idea of what follows from an angle-dependent specular parameter, we have plotted in figure 2 the conductivity for different constant parameters p_F and corresponding values of the quantity $q \equiv \frac{2}{3}\pi N_s S_0$ (cf. equation (11)), chosen so as to obtain the same asymptotic behaviour for thick films, $d/l \gg 1$, cf. (26) and (27). As the thickness becomes smaller than the MFP, Fuchs's conductivity decreases faster. This behaviour can be understood by keeping in mind the fact that an angular dependence of the form (11) renders ineffective the scattering of particles running almost parallel to the surface. Furthermore, as considered above, even their contribution gains importance for thin films.

The limiting expression (26) has a very appealing structure: the roughness-induced resistivity contribution, ρ_s , is proportional to the number of scattering centres per unit area

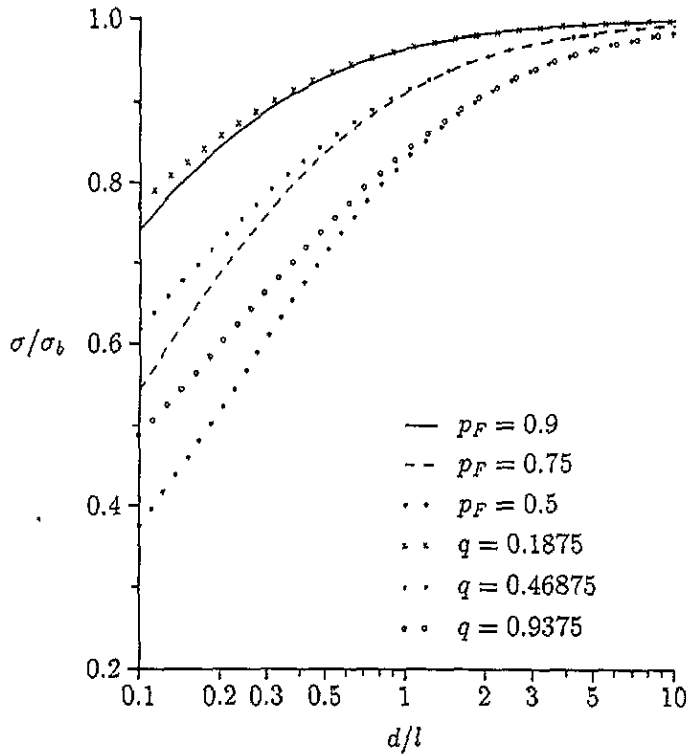


Figure 2. The reduced conductivity, σ/σ_b , is shown as a function of the ratio of the thickness to the mean free path, σ_b is Drude's bulk conductivity and corresponds to the diffusion constant D_b . The curves are calculated from equation (25) for three values of a constant specularity parameter, p_F , and the quantity q describing the fraction of diffusely scattered particles (11).

of the film surface and their corresponding scattering cross section, $\rho_s \simeq (l\sigma_b^{-1}) N_s S_0/d$. The quantity N_s/d can be viewed as the (volume) impurity density resulting from a distribution of the surface scatterers over the cross section. Consequently, $l_s \simeq d/N_s S_0$ may be defined as the MFP associated with surface-roughness scattering and thus, as usual for bulk scatterers, $\rho_s \simeq l_s^{-1}$. Generally, such a picture is known as the scattering hypothesis; it is discussed with respect to thin films by Wißmann [3]. Hence, we have confirmed a specific case of this hypothesis and given a derivation based on a microscopic model.

We have seen that the grazing incidence divergence of the conductivity can be prevented either by bulk scattering or by the discreteness of the lateral eigenvalues in a quantum mechanical framework. Each of them determines an upper limit θ_{\max} for oblique incidence. In particular, from the exponential attenuation $e^{(-d/l\zeta)}$ it follows, for $l > d$, $\theta_{\max} \simeq \cos^{-1}(d/l)$. The corresponding value due to a minimal lateral velocity component is given by $\theta_{\max} \simeq \cos^{-1}(\lambda_F/2d)$ where λ_F denotes the Fermi wavelength of the carriers. The smaller of these two cut-off angles may be considered as the upper limit of integration in equation (25). For $d^2 > l\lambda_F$, no effect from the underlying discrete energy-level structure remains; the system shows purely classical behaviour. Otherwise, a finite conductivity originates mainly from quantum mechanical properties. The value of the quantity $d^2/l\lambda_F$ determines therefore the region where a classical picture becomes invalid and quantum mechanics has to be used.

We have already mentioned that the basic scattering law (10) may also be deduced

from a mathematical formulation of surface scattering given in [17], [18] and [14]. In these papers, scatterers are considered as 'bumps' on the surface. It is worthwhile noticing that this situation differs from the case when the scatterers are distributed close to it. In connection with size effects, the latter model, for spherically symmetric scatterers, has been discussed by Chu and Sorbello [19].

Guided by theoretical considerations or experimental data, a number of angle-dependent specularly parameters have been proposed [13, 20–23]. As does (11), these models have in common that they guarantee specular reflection for grazing incidence. Only Soffer's model seems to be widely applied. In comparison to this, however, the method explained in section 2 offers a natural way of determining the specularly parameter in terms of properties of the surface. In particular, note the problems arising in [21] from the normalization procedure.

6. Summary

It is the aim of this paper to consider anew the transport in thin films with rough surfaces and to clarify a number of long-standing problems involved. In particular, we start with an analysis of the scattering at surface scatterers and introduce their differential scattering cross section, $S(\nu', \nu)$. We have shown that the time-reversal symmetry fulfilled by $S(\nu', \nu)$ is the condition for the homogeneity of the lateral carrier distribution. This physically appealing property was postulated by Fuchs [1] and is required for his calculations to hold. The resulting specularly parameter is uniquely determined by current balance and shows in general an angular dependence determined by a specific choice of S . Moreover, provided that the latter quantity has a physically sensible form (cf the discussion following (10)), the specularly parameter is always angle dependent. This result conflicts with the common model of a constant parameter. In view of our investigation, an angle-independent specularly parameter seems to be an oversimplified model and too far from reality. Finally we mention that it is only the more complicated scattering law (10) and the corresponding specularly parameter (11) which allow us to make contact with the quantum mechanical result (28), i.e., they give an explanation of the quadratic dependence of the conductivity on the thickness.

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Appendix

In the ensemble of all non-interacting carriers, moving with different energies, the density of the diffusion problem reads

$$\varrho(\mathbf{r}) = n(E)f(E - \mu(\mathbf{r})) \quad \text{grad } \varrho \equiv \partial\varrho/\partial\mathbf{r} = n(E)(df/dE)(-\partial\mu/\partial\mathbf{r}) \quad (\text{A1})$$

where f is (now) the one-particle distribution function, $\mu(\mathbf{r})$ the varying chemical potential, and $n(E)$ denotes the density of states (per unit volume) of a homogeneous bulk system.

According to the Einstein equivalence, $-\partial\mu/\partial r$ is proportional to a driving force F , i.e., the overall density gradient can be replaced by a constant field of force. The electrical current density can now be written in terms of the diffusion current

$$\mathbf{j}_{el} = e \int dE \mathbf{j}(E) = eF \int dE n(E) D(E) \left(-\frac{df}{dE} \right) \equiv \frac{1}{e} \sigma F \quad (\text{A2})$$

where e denotes the elementary charge and σ the conductivity. For a degenerate electron gas with a sharp Fermi energy, E_F , (A2) reduces to $\sigma = e^2 n(E_F) D(E_F)$. D/D_b can thus be replaced by σ/σ_b as usual.

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