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Adaptation of the specularity-parameter concept to size-quantized thin metallic films

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Abstract. We calculate the electrical conductivity σ of thin films where volume as well as surface scattering processes may occur. The latter arise from surface roughness and will be described in terms of the specularity parameter (SP). This concept, originally introduced by Fuchs, is generalized to a quantum-mechanical approach via the derivation of an SP-dependent transition rate. Since various angle-dependent SPS can be deduced from a microscopical description of the surface profile, our theory accounts in a very general manner for the interrelation between surface properties and resulting conductivity. If surface scattering is the dominant effect, we find a power law $\sigma_s(d) \sim d^{\alpha}$, where d is the thickness. Different exponents α of about two to three result from small or large roughness correlation lengths, respectively, and are of relevance in view of experimental data, too. For the combined action of volume and weak surface scatterers the theory predicts a wide applicability of a conductivity formula of Fuchs–Sondheimer type. This is confirmed very well by experiments on extremely thin films revealing quantum size-induced oscillations of the conductivity superimposed on an overall behaviour $\sigma_s(d) \sim d$.

1. Introduction

The electrical conductivity of thin 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 l (MFP) in the unbounded bulk. The surface contribution to the resistivity is attributed to surface roughness and gains importance as the thickness of the film d approaches the MFP (classical size-effect regime) or, for even thinner films, as the thickness is comparable to the Fermi wavelength λ_F of the electrons (quantum size-effect regime). In correspondence to these different regimes, theoretical descriptions of the influence of the surface on the conductivity are given in the framework of classical physics or quantum mechanics, respectively. The most important classical model was developed by Fuchs in 1938 [1]. His investigation is 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. Thus, p varies from unity for completely specular reflection to zero for completely diffuse scattering. In this sense, the classical approach is not restricted to weak roughness.

The specularity parameter (SP) p introduced originally by Fuchs is independent of the carrier's angle of incidence θ, i.e., this phenomenological quantity contains no direct information about the microscopic scattering mechanism itself. Guided by theoretical

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considerations or experimental data, however, a number of angle-dependent SPs $p(\theta)$ have been proposed [2, 3, 4, 5, 6]. These parameters depend on the microscopic properties of the surface. For instance, in a geometrical description of roughness, the SP will be a function of the mean square root deviation h of the height of the surface irregularities and their correlation length ξ, see section 4. Alternatively, roughness can be represented by scatterers that are distributed randomly over the surface [7, 6], see also [8]. Of course, one can imagine further models that yield a parameter for reflection in the specular direction in terms of properties of the surface. So, the introduction of an SP represents a very general and variable concept. Despite its limitations it provides a simple way of understanding and an analytically compact formulation of the interrelation between the surface profile and the resulting surface-roughness scattering.

The confinement of electrons in ultrathin films gives rise to a discretization of energy levels and thus renders necessary a quantum-mechanical description [9, 10, 11]. Different approaches have been proposed in order to incorporate surface-roughness scattering [12, 13, 10, 14, 15, 7]. Generally, however, they are restricted to the case that roughness represents a small perturbation only. In this limiting case, the system of lateral modes or eigenfunctions remains unchanged and scattering leads only to non-zero transition rates, i.e., the calculation of these rates is the central problem for a quantum-mechanical theory to be solved. The quantum-mechanical investigations lead to a very pronounced dependence of the conductivity on the thickness and it is very likely that only these results can account for some experimental data in ultrathin films.

In this paper we show how the SP concept can be introduced into a quantum-mechanical formalism. The resulting theory renders possible a clear and largely analytical discussion of different limiting cases which are usually evaluated numerically only [12, 14, 15]. As an example, we study the behaviour of the conductivity in films with a weakly or a strongly correlated surface profile, i.e., $\xi k_F \ll 1$ and $\xi k_F \gg 1$, respectively, where k_F is the Fermi wavevector. Furthermore, parameters $p(\theta)$ already used to describe classical size-effect data can be employed beyond this regime. In this respect, our generalization may be useful for experimentalists. Finally, our theory explains a rather surprising experimental result: in experiments which have revealed quantum size-effect oscillations of the conductivity the overall behaviour of the conductivity has been satisfactorily fitted by the Fuchs–Sondheimer approximation for thick films [16, 17]. This asymptotic expression should be valid for $d \gg l$ only and yields for the surface part of the conductivity the relation $\sigma_s \sim d$. The fact that no deviations from this classical formula appear results from a relatively smooth surface profile and requires the simultaneous occurrence of both surface and volume scattering.

The introduction of an angle-dependent SP requires the derivation of a corresponding transition rate, or, in our language, damping quantity. For weak roughness, this can be done straightforwardly employing familiar classical ideas and the transition from a continuously varying angle of incidence to its discrete analogue. The discretization of the angle of incidence is physically reasonable but a heuristic procedure, of course. A more elaborate derivation can be given in terms of the Green functions in a thin film. Up to linear order in $(1 - p) \ll 1$, we find the same roughness-induced transition rate. Although our paper is restricted to this weak-roughness case, this Green-function approach may lead to a quantum-mechanical theory including strong surface scattering, too.

The outline of the paper is as follows. In section 2 we summarize some formulae for the damping quantities, the conductivity, etc which we have derived and discussed in a recent paper [11], including volume scattering only. Later on, roughness-induced damping is introduced. In section 3, we give only a semiclassical derivation of this quantity whereas the Green-function approach is outlined in the appendix. Some examples for angle-dependent

SPs are given in section 4. In section 5, the resulting behaviour of the conductivity is discussed qualitatively and numerical calculations are presented. Finally, section 6 is used for a summary.

2. Basic formulae—volume scattering

The propagation of electrons can be described by the one-particle Green function. In a film with smooth surfaces, the Green function obeys the differential equation

$$\left[\Delta + k^2(r)\right] G(r, r') = -\delta(r - r')\, G|_{\text{surface}} = 0 \tag{1}$$

where we have assumed hard-wall boundaries. k is the medium wave number. The presence of configurationally averaged volume scatterers gives rise to an imaginary part $\operatorname{Im} k^2 > 0$ and is responsible for the attenuation of the coherent wave field [18]. The loss quantity is related to the local density of states according to [19]

$$\operatorname{Im} k^2(r) = (4\pi/l) \operatorname{Im} G(r, r') \tag{2}$$

where l is the mean free path associated with volume scattering in the unbounded 3D bulk. $G(r, r')$ can be represented in its most general form as a double sum over all lateral modes of the film [20]. For sufficiently weak scattering (i.e. $k_F l \gg 1$), however, we may restrict ourselves to the diagonal elements only because the non-diagonal ones are negligible [19]. Thus, equation (1) is solved by

$$G(r, r') = \frac{\mathrm{i}}{2d} \sum_{n=1}^{\infty} \sin(\kappa_n z) \sin(\kappa_n z')\, H_0^{(1)}(\mu_n |R - R'|) \tag{3}$$

$$r \equiv (R, z) \qquad 0 \leqslant z \leqslant d.$$

The lateral wave number is given by $\kappa_n = n\pi/d$ and $H_0^{(1)}$ is the Hankel function of first kind and zeroth order [21]. While the general form of the (2D) in-plane propagation is fixed by $H_0^{(1)}$, the properties of the individual modes are determined by the corresponding wave numbers μ_n. We find for these quantities

$$\mu_n^2 = (k^2)_{nn} - \kappa_n^2 \tag{4}$$

$$(k^2)_{nn} = (2/d) \int_0^d \mathrm{d}z \;\; \sin(\kappa_n z)\, k^2(z) \sin(\kappa_n z).$$

In the weak-scattering limit, the real part of the matrix element $(k^2)_{nn}$ is approximated sufficiently by $\operatorname{Re}(k^2)_{nn} \approx k_F^2$ and thus $\operatorname{Re} \mu_n^2 \approx k_F^2 - \kappa_n^2$. The imaginary part $\operatorname{Im} \mu_n^2$ is the characteristic damping quantity (or, except for a factor $\hbar/m$, transition rate) for each subband. Inserting equation (2) into formula (4) we obtain

$$\operatorname{Im} \mu_n^2 = (\pi/dl) \sum_{m=1}^{\infty} (1 + \tfrac{1}{2}\delta_{mn}) \left[\Theta(\operatorname{Re} \mu_m^2) - \pi^{-1} \tan^{-1}(\operatorname{Im} \mu_m^2 / \operatorname{Re} \mu_m^2)\right] \tag{5}$$

where Θ is the step function. Formulae (5) constitute an infinite system of non-linear equations. As discussed in [11], its solution yields the damping quantities with self-consistent level broadening. If we neglect these smearing effects, $\tan^{-1}(\operatorname{Im} \mu_n^2 / \operatorname{Re} \mu_n^2) \to 0$, we find the usual result for volume-scattering-induced damping [9, 10]

$$\operatorname{Im} \mu_n^2 = (\pi/dl)(n_c + \tfrac{1}{2}) \qquad n \leqslant n_c \tag{6}$$

where $n_c = \operatorname{int}\left[k_F d/\pi\right]$ is the number of conducting modes. For the purpose of this paper, we will often use this simple approximation instead of the exact expression (5).

The real and imaginary parts of μ_n^2 determine entirely the electrical conductivity

$$\sigma(d) = \frac{e^2}{2\pi\hbar d} \sum_{n=1}^{n_c} \frac{\mathrm{Re}\,\mu_n^2}{\mathrm{Im}\,\mu_n^2}. \tag{7}$$

Inserting the expression for $\mathrm{Re}\,\mu_n^2$ and $\mathrm{Im}\,\mu_n^2$ from equation (6) into the latter formula and taking the limit of thick films, $n_c \sim d \to \infty$, we get the (Drude) conductivity $\sigma_b = e^2 k_F^2 l/3\pi^2\hbar$ for the bulk system.

3. Roughness-induced damping

We assume that volume and surface scattering are weak enough to affect independently the electron's propagation process. Then, the corresponding damping quantities (or transition rates) are additive, i.e., surface roughness merely gives rise to an additional term $\Delta\mathrm{Im}\,\mu_n^2$ on the right-hand side of (5) or (6), respectively. According to $\Delta\mathrm{Im}\,\mu_n^2 = k_F/l_n$, this roughness-induced quantity defines a specific MFP l_n for each individual mode. In a classical framework, a MFP similarly determined by surface scattering can be derived in terms of the SP as follows [22]. In a relatively smooth film, $(1 - p(\theta)) \ll 1$, a carrier can undergo a series of specular reflections until it is scattered diffusely at the surface. Each reflection occurs with probability $p(\theta)$. The angle of incidence (and reflection), i.e. the angle θ between the carrier velocity and the surface normal, varies from zero (normal incidence) to $\pi/2$ (grazing incidence). The probability for a path of length L with a series of i consecutive reflections is given by $P(L) = p^i(\theta)$. Because $i \approx L\cos\theta/d$ one obtains $P(L) = \exp(L\cos\theta \ln p(\theta)/d)$. Thus, as well known from volume scattering, $P(L)$ obeys an exponential law and defines via $P(L) \equiv \exp(-L/l(\theta))$ a MFP $l(\theta) = (d/\cos\theta)\ln p^{-1}(\theta)$.

In a quantum-mechanical picture, the classical angle of incidence θ has to be restricted to certain discrete values in accordance with the underlying discrete energy-level structure. Guided by the fact that the angle of incidence for a plane wave is given by the ratio of the normal component of the wavevector to its total length, we are led to the discretization rule

$$\cos\theta \longrightarrow \cos\theta_n = \kappa_n/k_F = n\pi/dk_F \tag{8}$$

and thus $l(\theta) \to l(\theta_n) = l_n$. Formula (8) is not only physically appealing but can be confirmed by better founded investigations, too, cf. the appendix.

Combining the results of this simple exercise we obtain for the wanted quantity

$$\Delta\mathrm{Im}\,\mu_n^2 = \frac{n\pi}{d^2} \ln p^{-1}(\theta_n) \approx \frac{n\pi}{d^2}(1 - p(\theta_n)). \tag{9}$$

For the assumed weak surface scattering, this expression is approximated by the term on the right-hand side. The roughness-induced damping quantity (9) renders possible the incorporation of different angle-dependent SPs in a uniform manner. For surface-dominated scattering, it determines directly the resulting conductivity. Generally, however, both surface and volume scattering contribute to the total damping and account for a finite conductivity. It is noteworthy that while their damping quantities simply add, the total resistivity is not additively composed of two separate contributions. This is a consequence of the explicit dependence of the roughness-induced damping (9) on the mode index n. Remember that the corresponding volume quantity (5) is at least approximately (cf. equation (6)) independent of n in agreement with the assumed isotropic volume scattering.

As already mentioned in the introduction, rough surfaces can be considered in a Green-function approach, too (see appendix). In the limit of weak surface scattering, the damping quantity (9) is confirmed by these calculations.

4. Some specularity parameters

To illustrate the application of formula (9) let us consider three different SPs, namely p = constant, $1 - p(\theta) \sim \cos\theta$ and $1 - p(\theta) \sim \cos^2\theta$. The constant SP has already been introduced by Fuchs [1] and deserves no comment. The next was derived in [6] and reads explicitly

$$p(\theta) = 1 - \frac{2\pi}{3} N S_\circ \cos\theta. \tag{10}$$

In this paper [6] roughness is modelled by surface scatterers, scattering cross-section $S_\circ$ each, that are distributed uniformly and uncorrelatedly with mean density N. The third SP can be found employing the model of a continuously corrugated surface that is described by a profile function $h(\boldsymbol{R})$. A small amplitude of height deviations requires $hk_F \ll 1$, $h^2 = \langle h(\boldsymbol{R})^2 \rangle$, where $\langle \cdots \rangle$ denotes the configurational average. In this limiting case, the scattering of waves at the surface profile $h(\boldsymbol{R})$ can be calculated by the well known perturbation method [23, 24, 25, 26]. This approach yields for the reflection coefficient for scattering in the specular direction the general formula [25]

$$p(\theta) = 1 - 4k_F h^2 \cos\theta \ \mathrm{Re} \int \mathrm{d}^2 q \ C(k_F^{\parallel} - q) \sqrt{k_F^2 - q^2} \tag{11}$$

where

$$C(\boldsymbol{q}) = \int \frac{\mathrm{d}^2 \boldsymbol{R}}{(2\pi)^2} \, \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}} C(\boldsymbol{R}) \tag{12}$$

is the Fourier transform of the correlation function $C(\boldsymbol{R}) = h^{-2}\langle h(\boldsymbol{R}')h(\boldsymbol{R}' + \boldsymbol{R}) \rangle$ and $\cos^2\theta = 1 - (k_F^{\parallel})^2/k_F^2$. To proceed, $C(\boldsymbol{R})$ has to be specified. A commonly used *ansatz* is $C(\boldsymbol{R}) = \exp(-R^2/\xi^2)$, i.e., the correlation function is approximated as Gaussian [25, 14]. ξ is the roughness correlation length. Inserting this *ansatz* in (12) and (11), the SP $p(\theta)$ can be calculated analytically in the limit of small or large ξ, respectively,

$$p(\theta) = 1 - \tfrac{2}{3} k_F^4 h^2 \xi^2 \cos\theta \qquad \xi k_F \ll 1 \tag{13a}$$

$$p(\theta) = 1 - (2k_F h)^2 \cos^2\theta \qquad \xi k_F \gg 1. \tag{13b}$$

The differences in p resulting from different values of ξ are enhanced for grazing incidence ($\theta \to \pi/2$). As regards metals, the range of medium or even large parameters ξk_F should be of interest.

In the long-wave limit, formula (13*a*), the electrons do not resolve the internal structure of the surface irregularities. The portion of diffusely scattered carriers is determined by the mean area $\sim \xi^2$ that a correlated region of the surface profile covers. Consequently, a further reduction of the rather small quantity ξ in equation (13*a*) causes the SP $p(\theta)$ to increase.

The SPs (13*a*) and (10) are both of type $1 - p(\theta) \sim \cos\theta$. This agreement is reasonable if we bear in mind that the latter one relies on the assumption of uncorrelated surface scatterers. A comparison of both results yields the simple relation $\pi^{-1} k_F^4 h^2 \xi^2 = N S_\circ$ between the parameters used. Thus, in the following we may refer likewise to either of them. Finally, we remark that the SP (13*b*) can be considered as an expansion of Soffer's result [4] $p(\theta) = \exp[-(2k_F h \cos\theta)^2]$ in the limit of small $hk_F \ll 1$.

5. Conductivity

At the beginning of this section we discuss qualitatively the influence of the different SPs on σ. For this purpose, the volume-scattering-induced damping quantity (5) is sufficiently approximated by expression (6). The SPs are included via formula (9).

$(a)\, p =$ constant. In this case the total damping is given by

$$\operatorname{Im}\mu_n^2 = \frac{\pi}{dl}(n_c + \tfrac{1}{2}) + \frac{n\pi}{d^2}(1-p). \tag{14}$$

Substituting this expression into equation (7), we find for the film conductivity

$$\frac{\sigma(d)}{\sigma_b} = \frac{3}{2}\sum_{n=1}^{n_c} \frac{1 - n^2/x^2}{n_c + \frac{1}{2} + (l/d)(1-p)n} \xrightarrow{d \gg l} 1 - \frac{3l}{8d}(1-p) \tag{15}$$

where $x \equiv k_F d/\pi$. For thick films, $n_c \gg 1$, the summation in formula (15) can be replaced by an integration leading to the asymptotic expression on the right-hand side. Obviously, this is just the well known Fuchs–Sondheimer approximation. Thus, our quantum-mechanical approach reproduces exactly the corresponding classical result in the limit of a large number of conducting modes when the discretization of the energy levels can be neglected.

For vanishing volume scattering, $l \to \infty$, the surface-dominated conductivity is determined by the remaining term of equation (15), namely $\sum_n n^{-1}$. For $n_c \gg 1$, one gets $\sigma_s(d) \sim d \ln(dk_F)$. In a classical framework the Fermi wavelength is negligible compared with the film thickness so that we find the typical logarithmic singularity [1].

$(b)\, 1 - p(\theta) \sim \cos\theta$. This is the case of correlation lengths ξ which are small compared with k_F. The SP is explicitly given by equation (10) or (13a), respectively, leading to a roughness-induced damping $\Delta\operatorname{Im}\mu_n^2 \sim n^2/d^3$. Except for a slightly different choice of parameters, an identical rate has been found in [10]. The resulting conductivity attributable to surface roughness reads

$$\sigma_s(d) = \frac{3e^2}{4\pi\hbar k_F^3 h^2 \xi^2} x^2 \sum_{n=1}^{n_c} \frac{1 - n^2/x^2}{n^2} \sim d^2 \tag{16}$$

i.e., σ_s rises faster than in the case considered above. The dependence on the correlation length is given by $\sigma_s \sim \xi^{-2}$, cf. the discussion of the SP (13a) in the preceding section.

Generally, the conductivity is determined by the simultaneous occurrence of both volume and surface scattering. Then, for a large number of conducting modes, we find again a result of Fuchs–Sondheimer type

$$\frac{\sigma(d)}{\sigma_b} \approx 1 - \frac{2}{15} k_F^4 h^2 \xi^2 \frac{l}{d} \qquad d \gg l. \tag{17}$$

Comparing the asymptotic formulae (15) (RHS) and (17) one can identify the quantity $(16/45)k_F^4 h^2 \xi^2$ with the fraction $1 - p$ of diffusely scattered carriers in the Fuchs theory. This agrees with the result of Rodewald and Appel (formula (23) in [27]). Alternatively, employing the model of scatterers that are distributed on the surface, the constant SP can be expressed in terms of the quantity NS_0 according to $1 - p = (16\pi/45)NS_0 \approx NS_0$. This equation confirms previous results by Lenk and Knäbchen [6] and Kunze [7].

$(c)\, 1 - p(\theta) \sim \cos^2\theta$. The remaining case is characteristic for large correlation lengths $k_F\xi \gg 1$. Here, we obtain an even more pronounced dependence of the surface-dominated conductivity on the thickness, namely $\sigma_s \sim d^3$. On the other hand, the combined action of surface and volume scatterers leads to

$$\frac{\sigma(d)}{\sigma_b} \approx 1 - \frac{1}{2} k_F^2 h^2 \frac{l}{d} \qquad d \gg l. \tag{18}$$

This result is again of Fuchs–Sondheimer type.

According to the found interrelation between the SP and the corresponding surface-dominated conductivity ($1 - p(\theta) \sim \cos^{\alpha-1}\theta$ leads to $\sigma_s(d) \sim d^\alpha$), we can conclude that the smoother the surface is, the stronger rises σ_s with increasing thickness. On principle, even higher power laws than $\sigma_s(d) \sim d^3$ might occur for good-quality films. This is in agreement with experimental data, cf. references in [7]. However, we have to bear in mind that these power laws are restricted by volume scattering to a narrow region of very small d. In other words, as the surface influence decreases rapidly the volume contribution to the resistivity becomes dominant, i.e., one has to study σ instead of σ_s. The general form of σ can be written as

$$\frac{\sigma(d)}{\sigma_b} = \frac{3}{2x}\sum_{n=1}^{n_c}\frac{1-n^2/x^2}{1+c(l/d)(n/x)^\alpha} \tag{19}$$

where $n_c + \frac{1}{2}$ from equation (6) is approximated by x and $c < 1$ denotes the various prefactors belonging to the SPs. α runs from one to three for the SPs (14), (10) or (13*a*) and (13*b*), respectively. Formula (19) allows a simple evaluation in the case of thick films ($l/d \ll 1$) when its denominator, as a quantity close to unity, can be expanded. In this way, all asymptotic expressions ((15), (17) and (18)) have been derived.

Independent of l/d, the sum (19) is dominated by terms with large numerators, i.e., only lower modes n contribute significantly. Classically, these contributions can be assigned to particles moving almost parallel to the surfaces ($\theta \lesssim \pi/2$). Because of the small normal component of their wavevector they are very sensitive to the surface profile. This property is retained in the analytical description because a larger portion of specular reflection (a higher value of α) results in a drastically reduced factor $(n/x)^\alpha$ for these carriers. So, the denominator in equation (19) remains, even for $l \gtrsim d$, a quantity close to unity justifying the aforementioned expansion. Again, this procedure leads to formulae of Fuchs–Sondheimer type. We find therefore that for very smooth films with a highly correlated surface profile, the quantum-mechanically calculated conductivity is well approximated by the classical asymptotic expression even if the ratio l/d is not small compared to unity. Indeed, a broad applicability of the Fuchs–Sondheimer approximation has just been found in experiments where the appearance of quantum size-induced oscillations of the conductivity gives rise to the assumption of high-quality layers [16, 17]. The validity of the classical approximation is proved by measuring a surface conductivity $\sigma_s(d) = [\sigma^{-1}(d) - \sigma_b^{-1}]^{-1} \sim d$.

At the end of this section let us discuss some results calculated numerically, see figure 1. Now, the volume-induced damping is described by the exact formula (5) including both propagating and evanescent modes as well as level-broadening effects, cf. [11]. Up to this point, however, the damping (9) attributed to surface roughness was merely used for the propagating modes $n \leqslant n_c$ only. To broaden its range of applicability we have to generalize the discretization rule (8) to modes $n > n_c$. An evanescent wavefunction has only a propagating component normal to the surface. Accordingly, we have assumed that $\theta_n = 0$ (normal incidence) can be assigned to all of these modes. Of course, this is a purely heuristic reasoning but for the sake of brevity we should restrict ourselves to this argument. It can be detailed that this rule is mathematically consistent, too ([28] and cf. the appendix).

Now, taking into account this generalization of equation (8) we can form a total damping quantity $\operatorname{Im}\mu_n^2$ comprising the volume terms (5) and a roughness contribution. For the SP (10), the resulting system of equations has been solved numerically. Then, according to formula (7), the obtained values for $\operatorname{Im}\mu_n^2$ determine the conductivity which is plotted in figure 1. As can be seen roughness changes considerably the behaviour of σ for a small

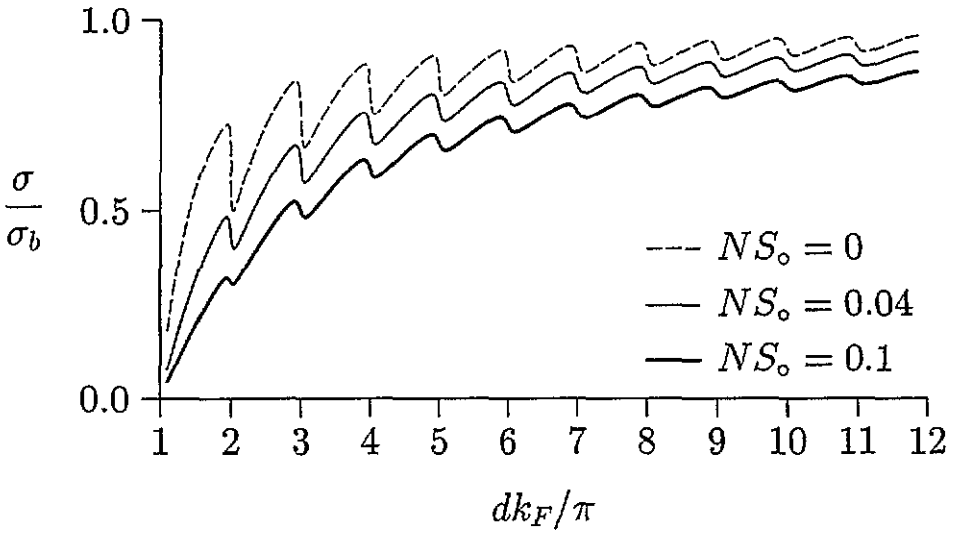

Figure 1. The reduced conductivity (7) is shown for a smooth and two rough layers. The angle-dependent specularity parameter is given by equation (10) and includes either the roughness parameter NS_o or, equivalently, the quantity $\pi^{-1}k_F^4h^2\xi^2$, cf. section 4. The damping due to volume scatterers is determined by the parameter $lk_F = 90$.

number of conducting modes whereas the curves are only shifted below that of a smooth layer ($NS_o = 0$) for higher values of n_c. In addition, the magnitude of the quantum size-induced oscillations is reduced. This effect is associated with level broadening (included in equation (5)) which is strongly enhanced for increasing scattering processes. In this respect, figure 1 confirms results found in a previous paper on volume scattering in thin films [11], namely that level broadening effects may drastically suppress the oscillatory behaviour of the conductivity, i.e., the clear manifestation of the quantum size effect.

6. Summary

It is the aim of this paper to put forward a basically quantum-mechanical theory of the conductivity in thin films where volume- as well as surface-scattering processes are taken into account. In particular, the latter are described by use of Fuchs's classical specularity parameter (SP). The main problem to be solved is the calculation of a corresponding, i.e. SP-dependent, damping quantity. Two separate derivations have been given, see section 3 and the appendix, respectively, in order to confirm the damping quantity used here. Nevertheless, it is obvious that this kind of approach involves some heuristic or semiclassical arguments. On the other hand, our theory has the advantage that, via the SP, different characterizations of surface roughness can be used straightforwardly in calculating the conductivity. In section 4 we have summarized, e.g., how to derive an SP from a geometrical description of the surface profile. One can introduce other SPs, too, or, in view of section 4, just employ a different roughness correlation function $C(R)$ to find further parameters $p(\theta)$. By virtue of this freedom, the theory provides a relatively comprehensive understanding of the interrelation between surface properties and resulting conductivity. So, we find for the surface-dominated conductivity the typical power law $\sigma_s(d) \sim d^\alpha$. The exponent α depends on the quality of the film: the smoother the surface is, the larger is α, i.e., the faster rises the conductivity with increasing thickness. For the more realistic, i.e. angle-dependent SPs

employed in this paper, α varies from two for uncorrelated or weakly correlated surface irregularities ($k_F\xi \ll 1$) to three for surface profiles exhibiting a rather large correlation length ($k_F\xi \gg 1$). In accordance with this range, a number of experimental data are available that suggest $\alpha \approx 2$. For $CoSi_2$ layers, e.g., Badoz *et al* [29] and Henz *et al* [30] have found $\sigma_s(d) \sim d^{2.3}$ and $\sigma_s(d) \sim d^{1.9}$, respectively. For thin Sn films, Orr *et al* [31] reported $\sigma_s \sim d^2$ (with a transition to $\sigma \sim d$ for larger thicknesses, cf. below).

A different behaviour $\sigma(d)$ arises from the combined action of surface and volume scatterers. Here, we have shown that weak surface scattering leads to a conductivity of Fuchs–Sondheimer type even if the condition this classical approximation is actually based on is not fulfilled. Accordingly, the formula $\sigma/\sigma_b = 1 - \text{constant} \times l/d$, similar to the original Fuchs–Sondheimer expression, should be widely applicable in experiments where films with a very smooth surface structure are grown [32]. In this sense, the occurrence of quantum size-effect oscillations of the conductivity may reasonably be accompanied by a mean conductivity obeying the classical asymptotic behaviour. Indeed, a quantum size-induced oscillatory behaviour of σ has been reported in experiments [17, 16] where the data could satisfactorily fitted by the Fuchs–Sondheimer approximation. For instance, the measurements by Jałochowski *et al* [16] show that also the conductivity for extremely thin films (from 0.1 to about 1 nm) agrees with this classical formula if modulations due to the quantum size effect and a periodically varying roughness (resulting from the layer-by-layer growth mode) are taken into account. This compatibility of the classical approximation with quantum size effects is strange at first glance and, to our knowledge, has not yet been discussed theoretically.

A conductivity of Fuchs–Sondheimer type is fixed by a single roughness parameter. In the original theory it is given by the phenomenological constant $1 - p$, see the right-hand side of equation (15). Employing the angle-dependent SPs (15) we have derived the formulae (17) and (18), respectively, in which this parameter is determined by more realistic quantities characterizing the microscopic surface profile. Besides this improvement itself, these formulae have the advantage that the used quantities are not subject to the restriction in Fuchs's model where $0 \leqslant 1 - p \leqslant 1$ has to be fulfilled. As a consequence, data giving rise to negative p values in the classical theory [16, 33, 34, 35] can be described by applying the proposed one, and values of the microscopic parameters can be extracted.

In concluding this paper we remark that the Green-function approach outlined in the appendix may be used for a theory that is not restricted to weak roughness only. Calculations along these lines will be left for the future.

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Appendix. Green-function approach

Equation (3) gives only one representation for the Green function in a film. Equivalent formulae can be found. One of them, useful in view of the incorporation of an SP, can be derived by considering all paths that contribute to the propagation of waves from a source point r' to r. The real paths include an increasing number of reflections at the surfaces but may be substituted by straight ones leading from a suitable chosen fictitious source point

outside the layer to r. This so-called image-potential method yields for the Green function the representation

$$G(r, r') = G_b(r, r') + \sum_{m=1}^{\infty}(-1)^m \left[G_m^+(r, r') + G_m^-(r, r')\right] \tag{A1}$$

with

$$G_m^{\pm}(r, r') = G_b(\sqrt{|R - R'|^2 + ((m \pm 1)d \mp z \mp z')^2}) \quad m \text{ odd}$$

$$G_m^{\pm}(r, r') = G_b(\sqrt{|R - R'|^2 + (md \mp z \pm z')^2}) \quad m \text{ even.}$$

The first term in equation (A1) describes the direct propagation from r' to r and is given by the 3D (bulk) Green function $G_b(r, r') = G_b(|r - r'|) = (4\pi|r - r'|)^{-1} \exp(ik|r - r'|)$. The sum in equation (A1) contains all processes with one or more specular reflections at the film boundaries. The indices $+$ and $-$ refer to the order of these reflections, namely a Green function labelled $+$ $(-)$ describes a propagation process with a last reflection at the upper (lower) surface $z = d$ $(z = 0)$. The additional factor $(-1)^m$ may be attributed to the phase change by π associated with each reflection. Of course, as does (3), the Green function (A1) obeys the defining equations (1).

Contrary to the original formula (3), the representation (A1) renders possible straightforwardly the use of an SP in order to describe the influence of statistically rough surfaces on the propagation process. According to Fuchs [1], roughness reduces the fraction of specularly reflected carriers in favour of a diffuse component. In his model, this redistribution is determined by the SP p. In our non-classical approach, the roughness-induced reduction of specular wave reflection can be introduced via an amplitude $\sqrt{p}$. A sequence of m reflections, in equation (A1) represented by $G_m^{\pm}$, gives rise to an attenuation factor $p^{m/2}$. Thus, we obtain

$$G(r, r') = G_b(r, r') + \sum_{m=1}^{\infty}(-1)^m p^{m/2} \left[G_m^+(r, r') + G_m^-(r, r')\right] \tag{A2}$$

for a Green function modified by surface roughness. The introduction of an angle-dependent SP $p(\theta)$ into equation (A2) is straightforward and merely requires us to weight the $G_m^{\pm}$ with individual factors $p^{m/2}(\theta_m^{\pm})$. For the sake of brevity, this generalized version is omitted.

One can show that (A2) and its generalization to $p = p(\theta)$ are consistently constructed in the classical limit when the discretization of the energy levels, the lateral dependence of the density of states etc are negligible. Indeed, employing a wave-superposition method, Fuchs's theory has been reproduced using the Green function (A2) [28].

Our formalism given in section 2 relies on the use of quantities which characterize the individual modes, cf., e.g., equation (7) for the conductivity. The further application of the propagator (A2) requires therefore the transition to a lateral-mode representation. For $p = 1$, these calculations must reproduce formula (3), of course. The deviations, however, resulting from a p value smaller than unity are more interesting.

The transformation of the Green function (A2) is essentially based on the so-called Sommerfeld integral [36]

$$4\pi G_b(r) = \frac{e^{ik\sqrt{R^2+z^2}}}{\sqrt{R^2+z^2}} = \frac{1}{2}\int_{-\infty}^{\infty} d\mu\ \mu\, H_0^{(1)}(\mu R)\, \frac{e^{-\tilde{\kappa}|z|}}{\tilde{\kappa}} \qquad \tilde{\kappa} = \sqrt{\mu^2 - k^2}. \tag{A3}$$

This identity separates the lateral component from the 2D (in-plane) propagation process described by the Hankel function $H_0^{(1)}$. Applying (A3) to all terms in equation (A2), one obtains

$$G(r,r') = \frac{1}{8d}\int_{-\infty}^{\infty} \mathrm{d}\mu\, \frac{\mu}{\tilde{\kappa}}\, H_0^{(1)}\left(\mu|R-R'|\right) L(z,z') \tag{A4}$$

$$L(z,z') = -\frac{\sqrt{p}}{1-p\,\mathrm{e}^{-2\tilde{\kappa}d}}\left[\mathrm{e}^{-\tilde{\kappa}(z+z')} + \mathrm{e}^{-\tilde{\kappa}(2d-z-z')}\right] + \frac{1}{1-p\,\mathrm{e}^{-2\tilde{\kappa}d}}\left[\mathrm{e}^{-\tilde{\kappa}|z-z'|} + p\,\mathrm{e}^{-\tilde{\kappa}(2d-|z-z'|)}\right].$$

The function $L(z,z')$ arises from the summation of all lateral contributions forming a geometrical series. Via the residue theorem, the integral over μ can be evaluated by exploiting the fact that L has poles at

$$\tilde{\kappa}_n = -\mathrm{i}\frac{n\pi}{d} + \frac{1}{2d}\ln p \qquad \text{for} \quad n = 0, \pm 1, \pm 2, \ldots. \tag{A5}$$

Because $\mu = \sqrt{k^2+\tilde{\kappa}^2}$, there are two complex roots μ_n for each pole $\tilde{\kappa}_n$. A single-valued correspondence is achieved by introducing a branch cut into the complex $\tilde{\kappa}_n^2$ plane. This cut has to connect the two branch points $\tilde{\kappa}_n^2 = -k^2$ and $\tilde{\kappa}_n^2 = \infty$ but can, in principle, be chosen arbitrarily. The favourable branch cut runs from point $-k^2$ along the negative real axis to infinity. This choice yields roots with a real part $\mathrm{Re}\,\mu_n > 0$, i.e. outgoing waves $H_0^{(1)}(\mu_n|R-R'|)$, and guarantees that the branch cut does not interfere with the poles $\tilde{\kappa}_n^2 = -(n\pi/d)^2 + ((\ln p)/2d)^2 - \mathrm{i}n\pi(\ln p)/d^2$. The points $\tilde{\kappa}_n^2$ form a parabola with the apex ($n = 0$) on the positive real axis and extending ($n \to \pm\infty$) to the left half space $\mathrm{Re}\,\tilde{\kappa}_n^2 < 0$.

The integration in equation (A4) can be performed along a contour which is closed in the upper half space where $H_0^{(1)}$ vanishes exponentially. According to the residue theorem, the value of this integral is determined by the enclosed poles. The final result reads

$$G(r,r') = \frac{\mathrm{i}}{2d}\sum_{n=1}^{\infty} H_0^1\left(\sqrt{k^2-\kappa_n^2}|R-R'|\right)\begin{cases}\cos\kappa_n(z-d/2)\cos\kappa_n(z'-d/2) & n \text{ odd}\\ \sin\kappa_n(z-d/2)\sin\kappa_n(z'-d/2) & n \text{ even}\end{cases} \tag{A6}$$

where $\kappa_n \equiv \mathrm{i}\tilde{\kappa}_n = n\pi/d + (\mathrm{i}/2d)\ln p$. As can be seen from a comparison with equation (3), roughness changes the lateral term of the Green function. Additionally, complex values κ_n occur. The total damping for each individual mode is given by $\mathrm{Im}\,\mu_n^2 = \mathrm{Im}\,(k^2-\kappa_n^2)$, i.e., we find the same roughness-induced damping

$$-\mathrm{Im}\,\kappa_n^2 \equiv \Delta\mathrm{Im}\,\mu_n^2 = -\frac{n\pi}{d^2}\ln p \tag{A7}$$

as in section 3, cf. formula (9).

In the weak-roughness regime, $1-p \ll 1$, the small imaginary part of κ_n is of importance for $\mathrm{Im}\,\kappa_n^2$ only. The quantity $\mathrm{Re}\,\mu_n^2$ is not changed since its additional term $\sim (\ln p)^2 \sim (1-p)^2$ is of higher order. Also the modified lateral functions in equation (A6) do not influence the conductivity (7) [28]. The derivation of equation (7), based on the formula (3), is given in [11] and can be repeated with the Green function (A6). For $1-p \ll 1$, these calculations confirm the conductivity (7) used in this paper. So, in linear order of $1-p$, the Green function (A6) yields only an enhancement of the intersubband transitions whereas the general formalism is not affected. This agrees exactly with the results one would expect from a first-order perturbational theory.

Finally, let us comment on the use of angle-dependent SPs. As noted above, our starting equation (A2) is readily generalized to functions $p = p(\theta)$. The appearance of individual prefactors $p^{m/2}(\theta_m^{\pm})$ renders ineffective the application of (A3). However, assuming that the SP is given as a function of the cosine of the angle of incidence, $p = p(\cos\theta)$, a generalized identity can be established

$$p\left(\frac{z}{\sqrt{R^2+z^2}}\right)\frac{e^{ik\sqrt{R^2+z^2}}}{\sqrt{R^2+z^2}} \approx \frac{1}{2}\int_{-\infty}^{\infty} d\mu\ \mu\, H_0^{(1)}(\mu R)\,\frac{e^{-\tilde{\kappa}|z|}}{\tilde{\kappa}}\,p\left(\frac{i\tilde{\kappa}}{k}\right). \qquad \text{(A8)}$$

On the left-hand side, the cosine is determined in real space. On the right-hand side, it is expressed by quantities in momentum space, namely the ratio of the normal component of the wavevector to the total wavevector. Mathematically, formula (A8) is confirmed by the method of steepest descent. This method is a standard technique in evaluating integrals with Hankel functions [36, 37] and, therefore, the explicit calculations can be omitted. Its application relies on the substitution of $H_0^{(1)}$ by the asymptotic expression $\sqrt{2/\pi\mu R}\exp[i(\mu R - \pi/4)]$ which is strictly justified in the limit of large arguments $|\mu R| \gg 1$ only. In this sense, the method of steepest descent can only yield a partial proof of equation (A8). We believe, however, that (A8) is sufficiently precise beyond that. (Even for $p =$ constant, when (A8) simplifies to the identity (A3), only a limited validity of formula (A8) would be anticipated from calculations with the method of steepest descent.)

Applying formula (A8) to the Green function (A2) with an angle-dependent SP, equation (A4) is re-found except that p is substituted by $p(i\tilde{\kappa}/k)$. The evaluation of this equation can be performed as described above. This procedure generalizes the roughness-induced damping (A7) to angle-dependent SPs and confirms the naive discretization rule (8). For $1 - p(\theta) \ll 1$, no further changes, compared to the case $p =$ constant, arise.

References

[1] Fuchs K 1938 *Proc. Camb. Phil. Soc.* **34** 100
[2] Ziman J M 1969 *Electrons and Phonons* (London: Oxford University Press) pp 452–60
[3] Parott J E 1965 *Proc. Phys. Soc.* **85** 1143
[4] Soffer S B 1967 *J. Appl. Phys.* **38** 1710
[5] Andreev A F 1971 *Sov. Phys.–Usp.* **14** 609
[6] Lenk R and Knäbchen A 1993 *J. Phys.: Condens. Matter* **5** 6563
[7] Kunze C 1993 *Solid State Commun.* **87** 359
[8] Schumacher D 1993 *Surface Scattering Experiments with Conduction Electrons (Springer Tracts in Modern Physics 128)* (Berlin: Springer)
[9] Sandomirskii V B 1967 *Zh. Eksp. Teor. Fiz.* **52** 158 (Engl. Transl. 1967 *Sov. Phys.–JETP* **25** 101)
[10] Trivedi N and Ashcroft N W 1988 *Phys. Rev.* B **38** 12298
[11] Knäbchen A 1994 *Thin Solid Films* submitted
[12] Leung K M 1984 *Phys. Rev.* B **30** 647
[13] Tešanović Z, Jarić M V and Maekava S 1986 *Phys. Rev. Lett.* **57** 2760
[14] Fishman G and Calecki D 1989 *Phys. Rev. Lett.* **62** 1302
[15] Fishman G and Calecki D 1991 *Phys. Rev.* B **43** 11581
[16] Jałochowski M, Bauer E, Knoppe H and Lilienkamp G 1992 *Phys. Rev.* B **45** 13607
[17] Schumacher D and Stark D 1982 *Surf. Sci.* **123** 384
[18] Lenk R 1989 *Phys. Status Solidi* b **154** 733
[19] Lenk R 1990 *Phys. Status Solidi* b **162** 227
[20] Lenk R 1991 *Solid State Commun.* **79** 531
[21] Ryshik I M and Gradstein I S 1957 *Tables of Series, Products, and Integrals* (Berlin: Deutscher Verlag der Wissenschaften)
[22] Cottey A A 1967–68 *Thin Solid Films* **1** 297
[23] Bass F G and Fuks I M 1979 *Wave Scattering from Statistically Rough Surfaces* (Oxford: Pergamon)

[24] Lekner J 1987 *Theory of Reflection* (Dordrecht: Martinus Nijhoff)
[25] Ogilvy J A 1987 *Rep. Prog. Phys.* **50** 1553
Equation (11) is deduced from formula (7.3) in Ogilvy's paper.
[26] Voronovich A G 1994 *Wave Scattering from Rough Surfaces (Springer Series on Wave Phenomena 17)* (Berlin: Springer)
[27] Rodewald B and Appel J 1975 *Surf. Sci.* **49** 21
[28] Knäbchen A 1994 *PhD Dissertation* University of Technology Chemnitz–Zwickau, Institute for Physics
[29] Badoz P A, Arnaud d'Avitaya F and Rosencher E 1991 *Ann. Phys., Paris* **16** 623
[30] Henz J, Hugi J, Ospelt M and von Känel H 1990 *Surf. Sci.* **228** 9
[31] Orr B G, Jaeger H M, Goldman A M and Kuper C G 1985 *Phys. Rev.* B **32** 7586
[32] Jałochowski M and Bauer E 1988 *Phys. Rev.* B **38** 5272
[33] Chopra K L and Randlett M R 1967 *J. Appl. Phys.* **38** 3144
[34] Bennett H E, Bennett J M, Ashley E J and Motyka R J 1968 *Phys. Rev.* **165** 755
[35] Marsocci V A and Shue S S 1971 *J. Appl. Phys.* **42** 504
[36] Sommerfeld A 1966 *Vorlesungen über Theoretische Physik* 6th edn, vol 6 (Leipzig: Geest & Portig)
[37] Landau L D and Lifschitz E M 1966 *Hydrodynamik, Lehrbuch der Theoretischen Physik* vol 6 (Berlin: Akademie)