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INFLUENCE OF THE SPECULARITY PARAMETER P ON THE CLASSICAL SIZE-EFFECT IN THIN METALLIC FILMS

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The influence of the specularity parameter P on the electrical conductivity of thin metallic films is discussed for different electron scattering models. A comparison between the models themselves, a new theoretical approach and some experimental data is done.

ВЛИЯНИЕ ПАРАМЕТРА ОТРАЖЕНИЯ НА КЛАССИЧЕСКИЙ МАСШТАБНЫЙ ЭФФЕКТ В МЕТАЛЛИЧЕСКИХ ТОНКИХ ПЛЕНКАХ

В работе анализируется влияние параметра отражения P на электропроводность металлических тонких пленок для разных моделей рассеяния электронов. Проводится сравнение трех разных моделей с новым теоретическим подходом и экспериментальными данными.

I. INTRODUCTION

It is well understood that when the thickness of a thin metallic film is of the same order of magnitude as the electronic mean free path, the scattering of the conduction electrons from the film boundaries becomes a significant factor. This leads to the so-called classical size effect. The interpretation of electrical conductivity measurements of thin metallic films is usually satisfactory when the well-known Fuchs size effect theory [1] is applied.

Fuchs defined the specularity parameter P as the probability that an electron will be specularly reflected after impinging upon the film surface. The value of P lies therefore between zero and one. Lucas [2] has extended the Fuchs treatment

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taking into account the effect of different specularity parameters P_1 and P_2 for the upper and lower surfaces of the film, respectively. Juretschke [3] has shown that even when the boundaries of the film are different, it is possible to replace the parameters P_1 and P_2 by a single effective coefficient P_{eff}. His definition of P_{eff} differs somewhat from the result obtained by Cottey [4]. Parrott [5] and Cotti [6] have proposed a simplified angular dependence of the specularity parameter for rough surfaces. In their model the constant specularity parameter P is replaced by a step function of the angle which gives for the specularity parameter the value one if the angle of incidence measured from the surface normal exceeds a certain critical value Θ_c and is zero below this value. Ziman [7] has used a different definition of the specularity parameter for rough surfaces. He has considered P to be a function of the geometrical roughness of the surface under the assumption that the probability distribution of the height of surface asperities is Gaussian. Soffer [8] has extended the Ziman results to the case of some oblique incidence. Finally, Grendel [9] has derived an expression for the specularity parameter P taking into account the influence of the electron gas degeneracy. The purpose of this work is to compare the results relating to the Grendel theory and previous definitions of the parameter P by means of calculating the electrical resistivity of thin metallic films.

II. THE MODELS FOR THE SPECULARITY PARAMETER P

An expression for the electrical conductivity (resistivity) of thin metallic films was first derived by Fuchs and afterwards generalized by Sondheimer [10] using the free electron theory and the appropriate solution of the Boltzmann transport equation determined by the boundary conditions at the surfaces. They deduced that if the surface of the film scatters conduction electrons diffusely, the resistivity is increased above the bulk value and becomes a function of the film thickness. On the other hand, if the electrons are reflected from the surface specularly, the film resistivity is the same as that of the corresponding bulk material and is independent from the film thickness (providing that the Fermi surface is spherical). They obtained the following expression for the ratio of the film resistivity ϱ_f to bulk resistivity ϱ_b:

$$\frac{\varrho_f}{\varrho_b}=\left\{1-\frac{3}{2K}\int_0^1 \mathrm{d}x(x-x^3)(1-A)(1-P)(1-PA)^{-1}\right\}^{-1}, \qquad (1)$$

where K is the reduced thickness (i.e. the ratio of the film thickness a to the bulk electron mean free path l), $x=\cos\Theta$ with Θ being the angle of incidence of the electron wave vector measured from the surface normal, P is the specularity parameter, and $A=\exp(-K/x)$.

There are three basic models for the specularity parameter:

(1) *The Fuchs model.* P_F — a completely phenomenological parameter — is the

fraction of the conduction electrons, which are reflected specularly. It possesses constant values (independently of the angle of incidence) from zero to one.

(2) *The Parrott-Cotti model.* P_{PC} is a step function of the angle of incidence Θ, namely

$$P_{PC}(\Theta)=1 \quad \text{for} \quad \Theta_c<\Theta<\pi/2 \qquad (2)$$
$$P_{PC}(\Theta)=0 \quad \text{for} \quad 0<\Theta<\Theta_c.$$

The explanation of this choice is based on considering the interaction of the electrons with the surface potential. Electrons with a small angle of incidence can penetrate deeper into the surface potential as those which have a large angle of incidence and are more likely to be directionally randomized, thus yielding P equal to zero.

(3) *The Soffer model.* Soffer pointed out for oblique impacts that Ziman had not taken into account the necessity of conserving the net flow of electrons corresponding to the incident and the reflected waves. His treatment of the size effect slightly modifies the theory of Ziman. When the same assumptions are made about the correlation functions of the height of the surface asperities as in the Ziman theory, the specularity parameter P exhibits an angular dependence given by the formula

$$P_S(\cos\Theta)=\exp\left\{-\frac{16\pi^2\eta^2}{\lambda^2}\cos^2\Theta\right\}, \qquad (3)$$

where η is the r.m.s. of the height of surface asperities (i. e. the roughness of the film surface), λ is electronic wavelength, and Θ is the angle of incidence of the conduction electrons measured from the surface normal.

Recently, Grendel has corrected all the previous definitions of the specularity parameters by taking into account the degeneracy of the conduction electron gas. He shows that the effective specularity parameter which must be taken into consideration in the Fuchs-type boundary conditions to the Boltzmann equation is

$$P_G=P\,\frac{1-f_0}{1-P^2f_0} \quad \text{for a degenerated gas} \qquad (4)$$
$$P_G=P \quad \text{for a non-degenerated gas,}$$

where f_0 is the electronic equilibrium distribution function (Fermi-Dirac). In the formula (4) P denotes a specularity parameter calculated without considering the Fermi-Dirac statistics. In metals mostly the conduction electrons in the vicinity of the Fermi level participate in the transport so that P_G from (4) for a degenerated gas becomes $P_G=P/(2-P^2)$.

For most metals the parameter P in formula (1) (i.e. according to Grendel P_G) is practically equal to zero. Thus it is immaterial to distinguish between P and P_G. In certain cases, e.g. for semimetals or, say, highly polished gold, the parameter P may be non-zero. If we use Grendel's parameter P_G in formula (1) as a phenomenological constant, then there is no actual difference between the Fuchs and the Grendel theory. Nevertheless, if one calculates P quantum-mechanically, then P_G is to be inserted into the Fuchs formula.

III. COMPARISON OF MODELS

The differences between the three basic models are illustrated in Fig. 1 taken from [8]. It shows that the Fuchs model leads to a resistivity increase without limit

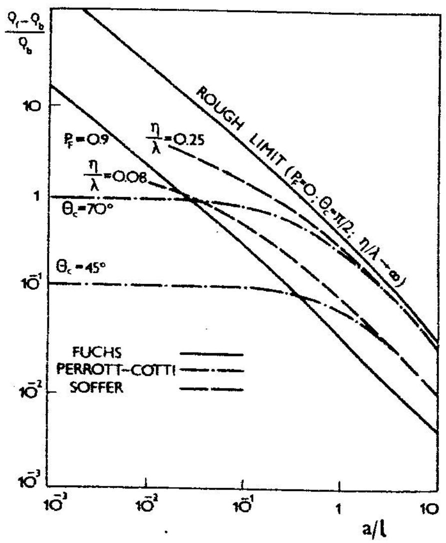

Fig. 1. The size effect in electrical resistivity according to the three different models.

as thickness is reduced unless the reflection is completely specular. This is due to the fact that on each reflection a fraction of $1-P$ electrons are lost to the current. The Parrott-Cotti model, on the other hand, gives a saturation in resistivity as the thickness is diminished, except in the case of $\Theta_c=\pi/2$. This is due to the fact that all electrons with $\Theta_c<\Theta<\pi/2$ are reflected specularly on successive reflections. The Soffer model leads to an intermediate behaviour. There is a small number of conduction electrons whose incidence is nearly grazing so that their specularity parameter, being near unity, reduces the size effect. All these three models however, are identical in the extreme rough or smooth limits.

Within the framework of the Grendel theory we have calculated the dependence of the film electrical resistivity on the reduced thickness K for various values of the surface roughness η (Grendel-Soffer) as well as for constant values of P (Grendel-Fuchs). A comparison between the previous results and those obtained from the Grendel theory is made below. Some experimental data are taken as references for the discussion.

Table 1 shows the average values of the Soffer specularity parameter P_S^{ave} and the corresponding average value of Grendel P_G^{ave} calculated as $P_i^{ave}=\int_0^{\pi/2} d\Theta \sin\Theta P_i(\cos\Theta)$, $i=S, G$, and $P_G=P_S/(2-P_S^2)$. Thus, P_i^{ave} is a value characteristic for most scattered electrons. The comparison of results according to Grendel and Soffer for various values η/λ is shown in Fig. 2. The curves show that for surface roughnesses $\eta>10\lambda$ the specularity parameter is for both the Grendel

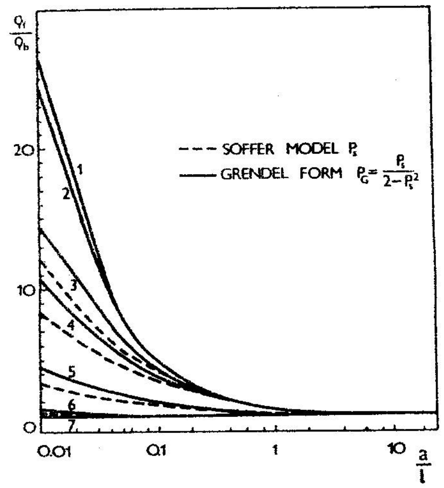

Fig. 2. The film resistivity versus the reduced thickness $K=a/l$. Note: 1, 2, ..., 7 refer to η/λ equal to ∞, 10, 1, 0.5, 0.1, 0.01, 0.

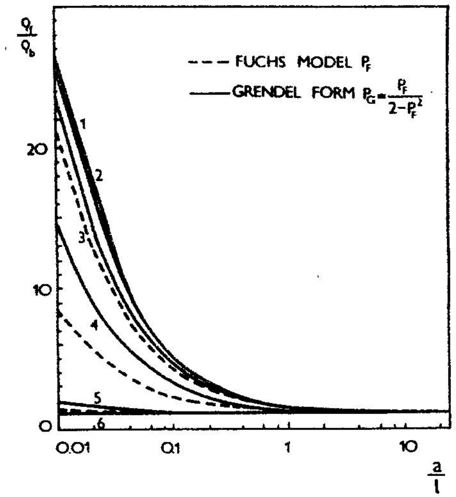

Fig. 3. The film resistivity versus the reduced thickness $K=a/l$. Note: 1, 2, ..., 6 refer to P_F equal to 0, 0.0074, 0.14, 0.65, 0.99, 1.

theory and the Soffer model almost always zero so that the reflection of the conduction electrons from the film surface will be completely diffuse. For surface roughnesses $\eta<0.01\lambda$ the specularity parameter in both cases is close to unity so that the reflection of the conduction electrons will be completely specular. For the surface roughness of the same order as the electron wavelength the average specularity parameter P_G^{ave} will be smaller than P_S^{ave}. Then the film electrical resistivity as a function of reduced thickness K calculated according to Grendel will possess higher values than the Soffer ones for the same roughness. (This is clearly seen for $K<1$.)

Table 1

The average values of P_s and $P_G = P_S/(2 - P_S^2)$ for some values of film roughness

η/λ	∞	10	1	0.5	0.1	0.01	0
P_S^{ave}	0	0.0074	0.07	0.14	0.65	0.99	1.0
P_G^{ave}	0	0.0056	0.05	0.10	0.50	0.98	1.0

Table 2

The Grendel specularity parameter P_G calculated for some constant values P_F

P_F	0	0.0074	0.07	0.14	0.65	0.99	1.0
P_G	0	0.0040	0.035	0.07	0.41	0.97	1.0

In the same manner a comparison between results according to Fuchs and Grendel is made. Table 2 shows the correspondence between the values of the Fuchs specularity parameter P_F (taking the same numerical values as P_S^{ave} of Table 1) and the Grendel values $P_G = P_F/(2 - P_F^2)$. The corresponding curves which illustrate the size effect are plotted in Fig. 3.

The comparison of calculations according to Grendel, Fuchs, and Soffer shows that the specularity parameter P is the same in both extreme cases, namely those of complete specular reflection ($P = 1$) and complete diffuse scattering ($P = 0$). It is also shown that for the specularity parameter $0 < P < 1$ the film electrical resistivity will be higher according to the Grendel theory (with the Soffer or the Fuchs P) than according to the Soffer or the Fuchs model alone, for the same values of the reduced thickness K.

IV. CONCLUSIONS

The original models for the specularity parameter P proposed by Fuchs, Parrott and Cotti, and Soffer agree satisfactorily with the experimental results in the thickness range of several micrometers. However, at smaller thicknesses some discrepancies between theory and experiment have been reported [11, 12].

Most, but not all, experimental results have been consistent with the Fuchs model with $P = 0$. Since all models and theories agree in the rough limit, this should not be taken as a verification of the Fuchs model. Only for the partially specular reflection the free electron models and theories can be distinguished by their different thickness dependences. For instance, Shimomura [13] has found that for thin gold films the value $P = 0.45$. If we attribute this value to P_G in the Grendel theory, the corresponding Fuchs model value is $P_F = 0.69$. Thus, P_G is smaller, i.e. the scattering is more diffuse — a simple consequence of the Pauli exclusion principle which causes a further directional randomization of the surface scattering.

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