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Analysis of the electrical resistivity of double-layer metal film

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Assuming that the various sources of scattering act independently in metal films and introducing the mean-free path describing the Fuchs–Sondheimer effect and the grain-boundary scattering, an apparent size effect is defined for a double-layer metal film; a similar method is used for representing any source of scattering with reference to the base layer. General expressions for the electrical conductivity of a double-layer metal film are thus obtained. Qualitative agreement with previous results of other authors is found in the thickness range where these early expressions are valid.

1. Introduction

Several authors [1–4] have analysed theoretically the effect of a metal overlayer on the electrical conductivity of a metal film; in all cases the analytical form of the total conductivity is sophisticated and the electronic scattering at the grain boundary is omitted.

As some experiments have shown [5] that a very thin overlayer could cause a drastic variation in the film conductivity, we attempt in this paper to interpret these phenomena in the framework of multidimensional conduction models [5–7] based on the representation of any scattering effect by a mean free path [5]. This has been shown [5–8] to be a convenient tool for interpreting the transport properties in the total extent of the experimental domain, i.e. whatever the film structure and the surface roughness e.g. annealed or unannealed; this first point has been recently emphasized [7].

2. Theory

2.1. General assumptions

Previous theoretical studies [8] lead to a unique form of analytical equation for representing a given transport property of a metal film, either resistivity or Hall coefficient or thermoelectric power or thermal conductivity; this type of equation holds when the film exhibits either a monocrystal or monocrystalline or columnar or polycrystalline structure.

Moreover, the existence of film surfaces having unlike properties is easily taken into account in the calculation of the transport properties in the frameworks of the two- and three-dimensional conduction models [5–8].

However, we must not forget that the following basic assumptions are used [5]:

(i) the different sources of scattering (phonons, grain-boundaries and external surfaces) act independently;

(ii) the effect of any scattering source may be represented by an associated electron mean free path;

(iii) the effect of the roughness of an external surface is represented by the electronic specular reflection coefficient, p, earlier introduced by Fuchs [9];

(iv) the statistical effect of the roughness of the grain-boundary is represented by a transmission coefficient, t [5, 10], in the way suggested by Cottey and Warkusz [11–13].

Assumption (ii) gives an alternative procedure [14] for calculating the Fuchs–Sondheimer size effect [15]; the numerical values of conductivity obtained under this assumption are close to the exact ones [14].

This feature had not been established by Cottey [11], who initiated this description of the scattering at external film surfaces, because he only considered the case of soft surfaces; further studies [5, 16–20] in this way have led to more general expressions [7, 8], valid for any value of p and gave a theoretical basis [14] for identifying the extension of the Cottey model [5, 16] with the Fuchs–Sondheimer model [15].

For a theoretical description of the effect of an overlayer the following further assumption is used:

(v) the top surface of the metal layer on which the overlayer is deposited is regarded as a grain boundary.

This assumption requires that the overlayer be continuous and does not allow the study of quantum size effects [5] for which special models have been proposed by several workers [5].

Let us remember that when two different specular reflection coefficients p_1 and p_2, must be used for describing external metal surfaces exhibiting unlike roughness properties, a unique effective coefficient, $\bar{p}$ [18, 21] may be introduced in the general equations obtained in the frameworks of the multidimensional models [5], with either [18]

$$\bar{p} = (p_1 p_2)^{1/2} \qquad p_1, p_2 \approx 1 \qquad (1a)$$

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or [22]

$$\bar{p} = [(1 + p_1)(1 + p_2) - (1 - p_1p_2)] \times [(1 + p_1)(1 + p_2) + (1 - p_1p_2)]^{-1} \quad (1b)$$

2.2. The apparent size effect

Introducing the transmission coefficient of the layer–overlayer boundary, s, and the electronic specular reflection coefficient at the top surface of the overlayer, q, it is clearly seen (Fig. 1) that the situation is similar to an external surface scattering with a resultant specular coefficient, p_r, given by

$$p_r = s^2q \quad (2)$$

provided that any electron path crosses the layer and the overlayer; this simplified view is acceptable when the electron mean free path in any bulk material is much larger than the overlayer thickness; this simplifying assumption is retained in this paper.

The apparent size effect can then be interpreted on the basis of the extended Cottey model [5, 20] by introducing the apparent specular reflection coefficient p_a, given by either

$$p_a = (p_rp_1)^{1/2} \qquad p_r, p_1 \approx 1 \quad (3a)$$

or

$$p_a = [(1 + p_1)(1 + p_r) - (1 - p_1p_r)] \times [(1 + p_1)(1 + p_r) + (1 - p_1p_r)]^{-1} \quad (3b)$$

where p_1 is the specular reflection coefficient at the base external surface of the metal film (Fig. 1).

Introducing Equation 2 into Equation 3, gives

$$p_a = (s^2qp_1)^{1/2} \qquad s^2q, p_1 \approx 1 \quad (4a)$$

$$p_a = [(1 + p_1)(1 + s^2q) - (1 - s^2qp_1)] \times [(1 + p_1)(1 + s^2q) + (1 - s^2qp_1)]^{-1} \quad (4b)$$

2.3. Background scattering

Given the thickness and the value of the electron mean free path in the material building the layer and the overlayer, d_1 and λ_{01} and d_2 and λ_{02}, respectively, and defining the relative thickness of the overlayer, m, by the equation

$$m = d_2/d_1 \quad (5)$$

and assuming that along any electron path the path lengths within the overlayer and the layer, L_1 and L_2 respectively, are related by Equation 5, the apparent

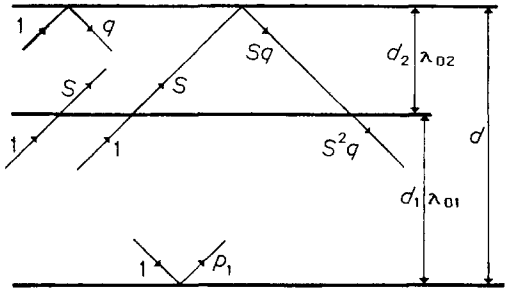

Figure 1 Electrical and geometrical parameters for the double-layer film.

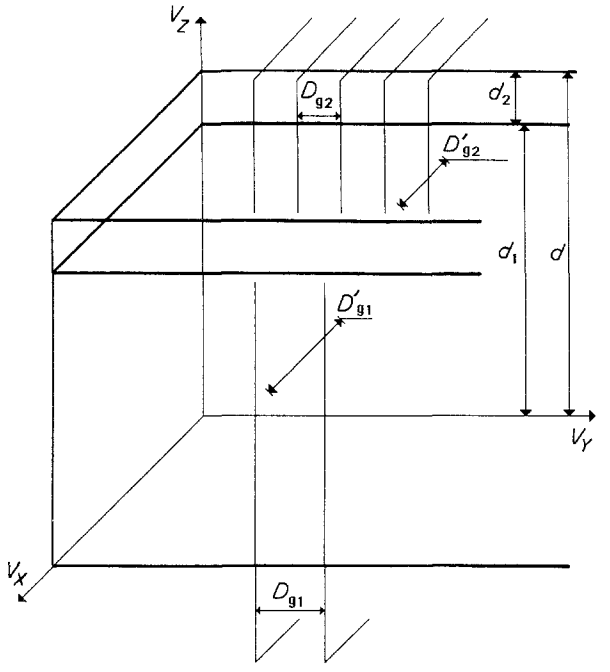

Figure 2 Definition of the interspace along a given electron path, D'_{g1} and D'_{g2}, in the case of an array of planar grain boundaries perpendicular to V_y.

probability that an electron carries along a path of length, L, before scattering, $p(L, \lambda_a)$, is expressed as

$$p(L, \lambda_a) = \exp\left(-\frac{L}{\lambda_a}\right) = \exp\left(-\frac{L_1}{\lambda_{01}} - \frac{L_2}{\lambda_{02}}\right) \quad (6)$$

where λ_a is the apparent bulk mean free path (Fig. 2).

Equation 6 can be rewritten as

$$p(L, \lambda_a) = \exp\left[\left(-\frac{L_1}{\lambda_{01}}\right)\left(1 + m\frac{\lambda_{01}}{\lambda_{02}}\right)\right] \quad (7)$$

Consequently

$$\lambda_a = \lambda_{01}\left[1 + m\frac{\lambda_{01}}{\lambda_{02}}\right]^{-1}(1 + m) \quad (8)$$

an alternative form of which is

$$\lambda_a = \lambda_{01}\left[1 + m\left(1 - \frac{\lambda_{01}}{\lambda_{02}}\right)\Big/\left(1 + m\frac{\lambda_{01}}{\lambda_{02}}\right)\right] \quad (9)$$

2.4. Grain-boundary scattering

In close similarity to the above assumptions and also assuming that the grain diameter in the layer and the overlayer is D_{g1} and D_{g2}, respectively, and that the arrays of grain boundaries of the layer and of the overlayer both extend in two or three dimensions, the number of intercepts, N, of an electron path at the arrays of grain boundaries is defined by

$$N = N_1 + N_2 \quad (10)$$

where

$$N_1 = L_1/D'_{g1} \quad (11a)$$

$$N_2 = L_2/D'_{g2} \quad (11b)$$

where D'_{g1} and D'_{g2} are the distances along the electron path between two successive intercepts with a given

array of grain boundaries (Fig. 2). Hence

$$N = \frac{L_1}{D'_{g1}}\left[1 + m\frac{D'_{g1}}{D'_{g2}}\right] \quad (12a)$$

$$N = \frac{L_1}{D'_{g1}}\left[1 + m\frac{D_{g1}}{D_{g2}}\right] \quad (12b)$$

It then seems convenient to define an apparent grain diameter, D_{ga}, defined by

$$N = \frac{L_1 + L_2}{D_{ga}} = \frac{L}{D_{ga}} \quad (13)$$

Equations 12 and 13 then give

$$D_{ga} = D'_{g1}\left[1 + m\frac{D_{g1}}{D_{g2}}\right]^{-1}(1 + m) \quad (14)$$

The whole set of the layer and thin overlayer (with respect to the bulk electron mean free path) can then be replaced by a fictive layer of thickness equal to the total film thickness, with an apparent grain diameter D_{ga} and an apparent mean free path λ_a, and whose surface roughness can be described by an apparent specular reflection coefficient, p_a.

In the usual equation [5] for the electrical conductivity in the framework of any multidimensional model [5, 20] the bulk conductivity of the layer, σ_{01}, may then be replaced by σ_{0a}, with

$$\sigma_{0a} = \frac{\sigma_{01}}{\lambda_{01}}\lambda_a = \sigma_{01}(1 + m)(1 + m\lambda_{01}\lambda_{02}^{-1})^{-1} \quad (15)$$

as derived from Equation 8.

2.5. Total film conductivity

The electrical conductivity of the total film, σ_f, is then [5–8]

$$\sigma_f = \sigma_{01}\lambda_a/\lambda_{01}A(b, \gamma) \quad (16)$$

with

$$b = \mu^{-1} + C_1\nu^{-1} \quad (17)$$

$$\gamma = b^{-1}(1 + C^2\nu^{-1}) \quad (18)$$

$$\mu = (d_1 + d_2)\lambda_a^{-1}(1 + p_a)[2(1 - p_a)]^{-1} \quad (19)$$

$$\nu = D_{ga}\lambda_a^{-1}(1 + t)[2(1 - t)]^{-1} \quad (20)$$

$$A(b, \gamma) = \frac{3}{2b}[\gamma - \tfrac{1}{2} + (1 - \gamma^2)\ln(1 + \gamma^{-1})] \quad (21)$$

where

$$C = 4/\pi \quad (22)$$

$$C_1 = 1 - C, \text{ for polycrystalline films} \quad (23a)$$

$$C_1 = -C, \text{ for monocrystalline and columnar films} \quad (23b)$$

In the case where the thickness of the overlayer is much thinner than that of the layer, asymptotic formulae can be used, as follows

$$\lambda_a \approx \lambda_{01}\left[1 + m\left(1 - \frac{\lambda_{01}}{\lambda_{02}}\right)\right] \quad (24)$$

$$D_{ga} \approx D'_{g1}\left[1 + m\left(1 - \frac{D_{g1}}{D_{g2}}\right)\right] \quad (25)$$

$$\sigma_{0a} \approx \sigma_{01}\left[1 + m\left(1 - \frac{\lambda_{01}}{\lambda_{02}}\right)\right] \quad (26)$$

$$\mu \approx d_1\lambda_{01}^{-1}\left[1 + m\frac{\lambda_{01}}{\lambda_{02}}\right](1 + p_a)[2(1 - p_a)]^{-1} \quad (27)$$

$$\nu \approx D_{g1}\lambda_{01}^{-1}\left[1 + m\left(-\frac{D_{g1}}{D_{g2}} + \frac{\lambda_{01}}{\lambda_{02}}\right)\right] \times (1 + t)[(2(1 - t)]^{-1} \quad (28)$$

When the overlayer is very thin, one can reasonably assume that the nucleation-growth procedure exhibits an epitaxial behaviour at the initial step, hence

$$D_{g2} \approx D_{g1} \quad (29)$$

and finally

$$\lambda_a \approx \lambda_{01}(1 + mu) \quad (30)$$

$$D_{ga} = D_{g1} \quad (31)$$

$$\sigma_{0a} = \sigma_{01}(1 + mu) \quad (32)$$

$$\mu \approx \mu_1[1 + m(1 - u)] \quad (33)$$

$$\nu \approx \nu_1[1 - mu] \quad (34)$$

with

$$u = 1 - \frac{\lambda_{01}}{\lambda_{02}} \quad (35)$$

$$\mu_1 = d_1\lambda_{01}^{-1}(1 + p_a)[2(1 - p_a)]^{-1} \quad (36)$$

$$\nu_1 = D_{g1}\lambda_{01}^{-1}(1 + t)[2(1 - t)]^{-1} \quad (37)$$

For a large range of film thickness, a recent study [8] has proposed a simple asymptotic equation for the reduced conductivity, i.e.

$$\sigma_f/\sigma_0 \approx (b\gamma + C_2 b)^{-1} \quad (38a)$$

with

$$C_2 = 3/8 \quad (38b)$$

Hence, from Equations 17, 33, 34 and 38, the following equation is derived:

$$\sigma_f/\sigma_{01} \approx (1 + mu)\left[1 + \frac{C^2}{\nu_1}\frac{1}{1 - mu} + C_2\frac{1}{\mu_1}\frac{1}{1 + m(1 - u)} + C_2\frac{C_1}{\mu_1}\frac{1}{1 - mu}\right]^{-1} \quad (39)$$

i.e.

$$\sigma_f/\sigma_{01} \approx \left(1 + \frac{C^2}{\nu_1} + \frac{C_2}{\mu_1} + \frac{C_1C_2}{\nu_1} - mu - m\frac{C_2}{\mu_1}\right)^{-1} \quad (40)$$

$$\sigma_f/\sigma_{01} \approx \left\{\sigma_1/\sigma_{01}\left[1 - m\frac{\sigma_{01}}{\sigma_1}\left(\frac{C_2}{\mu_1} + u\right)\right]\right\}^{-1} \quad (41)$$

with

$$\sigma_1/\sigma_{01} = \left(1 + \frac{C^2}{\nu_1} + \frac{C_2}{\mu_1} + \frac{C_1C_2}{\nu_1}\right)^{-1} \quad (42)$$

3. Comparison with previous theoretical works

Lucas [2] has proposed an analytical equation in the case of a double-layer metallic film exhibiting specular

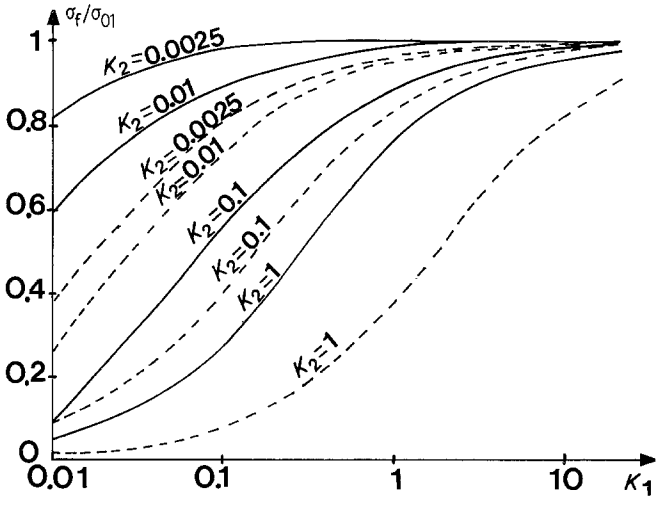

Figure 3 Variations in the reduced electrical conductivity in the framework of Lucas' Equation 16 [12] (——), and from Equation 16 (– – –).

scattering. The reduced film conductivity is written as follows ([2] Equation 16)

$$\sigma_f/\sigma_0 = (d_1 + d_2)^{-1}\left[d_1 + d_2\lambda_{02}\lambda_{01}^{-1} + \tfrac{3}{4}\lambda_{01}^{-1}(\lambda_{01} - \lambda_{02})^2 \int_0^1 F(T)\,dT\right] \quad (43)$$

where

$$F(T) = (T^3 - T)(1 - X^2)(1 - Y^2)[1 - X^2Y^2]^{-1} \quad (44)$$

with

$$X = \exp - K_1/T \quad (45a)$$

$$Y = \exp - K_2/T \quad (45b)$$

Variations in $-\int_0^1 F(T)\,dT$ with K_1 have been calculated by Lucas, for a series of values of K_2 ([2] Fig. 3). For the sake of simplicity, the values of σ_f/σ_0 have then been calculated (Equation 43) by introducing the numerical values of the integral $\int_0^1 F(T)\,dT$ into Equation 43 (Fig. 3).

Numerical values of Equation 16 have been calculated, neglecting the effect of grain boundaries ($D_g = \infty$) and assuming that the electronic scattering at the base layer is completely specular. The data corresponding to $p_a = 0.95$ are given in Fig. 3; this value leads to a minimum deviation from Lucas' results. For any value of p_a a qualitative agreement is observed.

However, our attention can be focused on the assumptions used by Lucas [2]. On the one hand, he assumed that the scattering at the external surfaces of the *double* layer is specular and calculated the double-layer conductivity from a weighted average of the conductivity of any layer ([2] Equation 14]; this assumption holds when perfectly specular scattering occurs at the layer interface; on the other hand, he writes continuity relations for the electron distribution functions at the interface ([2] Equation 9). These two assumptions are not physically consistent. Consequently, the marked deviation at low K_2 between Lucas' expression and the new equations cannot sustain any criticism against the proposed new model. Furthermore the variations in p_a with K_2 show that an overlayer could accentuate the roughness of the surface of the double layer, whereas the base layer exhibited smooth surfaces.

Bezak *et al.* [3] have proposed a general equation for the conductivity of a double-layer film starting from the Boltzmann equation and assuming that the specularity coefficients are given by Ziman–Soffer formulae and that the layer interface is represented by a potential step.

The analytical equation is somewhat complicated and numerical values of the reduced conductivity have been computed [3] in the case where the scattering at the lower film boundary is totally specular and at the top boundary is totally diffuse.

Neglecting grain-boundary scattering and taking the constant value 0.65 for p_a and introducing Equations 14 to 20 in Equation 38, leads to numerical data which are in qualitative agreement with Bezak *et al*'s numerical data (Fig. 4); a marked quantitative deviation occurs in low values of m.

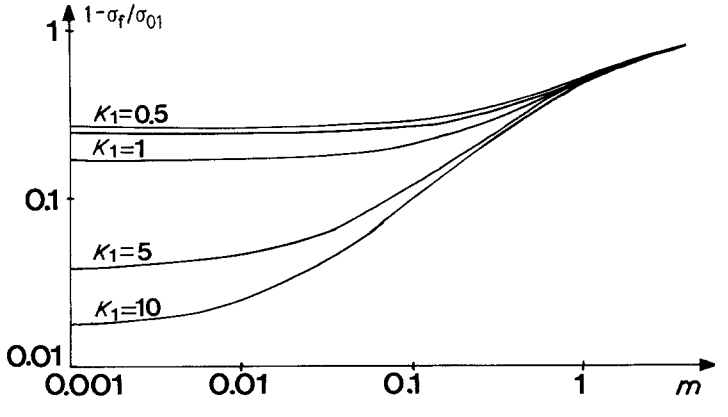

Figure 4 Variations in the reduced electrical conductivity in the framework of Bezak *et al.* (Equation 10, by taking $\lambda_{01}/\lambda_{02} = 10$) [3].

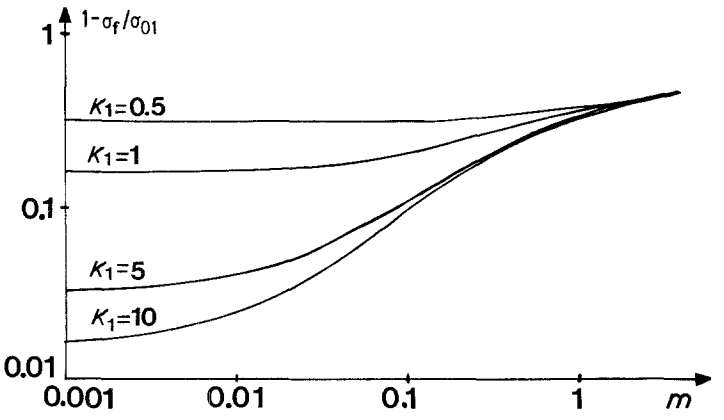

Figure 5 Variations in the reduced electrical conductivity in the framework of a multidimensional conduction model (Equations 14 to 20 in Equation 38, by taking $\lambda_{01}/\lambda_{02} = 2$).

At low values of m the results of Bezak *et al.* (related $\lambda_{01}/\lambda_{02} = 10$) can be obtained from the proposed equations by taking $\lambda_{01}/\lambda_{02} = 2$ (Fig. 5). An origin for this discrepancy could be found in the fact that Bezak *et al.* have written the conservation of energy for an electron travelling from layer 1 to layer 2 (Section 2.1 and Fig. 2) (hence $V_{F1} > V_{F2}$), neglecting the fact that the distribution of electrons in layer 1 also includes electrons which have been travelling from layer 2 to layer 1 that requires $V_{F2} > V_{F1}$ except if the scattering at the top surface is purely diffuse; consequently the calculated value of the conductivity is lower than the true one.

4. Conclusion

The analysis of the electrical resistivity of a double-layer metal film can be based on apparent background grain-boundary and film-surface scattering defined in the framework of multidimensional conduction models [5]. Qualitative agreement with the earlier works of Lucas [2] and Bezak *et al.* [3] is observed.

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