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Linear variations in conductivity with thickness of thin polycrystalline films

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Starting from a three-dimensional scattering model and from the mean free path representing the effect of electronic scattering on external surfaces, a linear analytic expression is proposed for thin film conductivity and resistivity. Good agreement with experiment is found.

1. Introduction

In order to take into account the effect of grain-boundary scattering on the transport properties of metal films, a three-dimensional scattering model has been recently proposed [1]; a transmission coefficient of grain boundaries was introduced for this purpose and it has been observed [2] that reasonable values of this coefficient could be deduced from data related to r.f. sputtered films [3–5].

It has also been shown [6, 7] that the Cottey conduction model [8] could be used to express the effect of electronic scattering at the external surfaces of the film. Hence in this paper we attempt to express the film conductivity in the presence of simultaneous background grain-boundary and external surfaces scattering by using the electronic mean free paths derived from the three-dimensional model [1] and from the Cottey conduction model [8].

2. Film conductivity

2.1. General theoretical expression

In a statistical three-dimensional model [1] the electronic mean free path λ related to the grain-boundary scattering can be expressed in polar co-ordinates (r, θ, ϕ) by:

$$\lambda^{-1} \approx D^{-1}(\ln 1/t)\{(C-1)\,|\sin\theta| + C\}, \quad (1)$$

where D is the average grain diameter, t the transmission coefficient (i.e. the fraction of electrons which are specularly transmitted through any grain boundary), and C a constant, whose value is $4/\pi$ [1].

Starting from the Cottey's analysis of scattering at external surfaces, a mean free path, λ_S, can be defined; λ_S is calculated in the same way as λ [1, 9] by substituting d for D and p for t:

$$\lambda_S^{-1} = d^{-1}\cdot(\ln 1/p)\cdot|\cos\theta|, \quad (2)$$

where d is the film thickness and p the usual electronic specular reflection coefficient.

Assuming that the three types of scattering give separate effects, the resulting mean free path, l, is given by:

$$l^{-1}(\theta,\phi) \approx l_0^{-1} + \lambda^{-1} + \lambda_S^{-1}, \quad (3)$$

where l_0 is the bulk mean free path.

In order to express the film conductivity, σ_f, the expression of l is introduced in the well-known Equation 4 [8, 10, 11], which is derived from the Boltzmann formulation of the distribution function of electrons, the electric field being applied in the X-direction:

$$\sigma_f = 2e\left(\frac{m}{h}\right)^3 V^2 l_0 \int_0^{2\pi} d\phi \int_0^{\pi} \frac{\sin^3\theta\,\cos^2\phi}{1 + l_0\cdot l^{-1}}\,d\theta. \quad (4)$$

Defining the grain parameter ν and the film parameter μ as:

$$\nu = l_0^{-1}\cdot D\cdot\left(\ln\frac{1}{t}\right)^{-1} \quad (5)$$

$$\mu = l_0^{-1}\cdot d\cdot\left(\ln\frac{1}{p}\right)^{-1}; \quad (6)$$

introducing them into Equations 1, 2 and 4 and finally integrating Equation 4 gives:

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$$\sigma_f/\sigma_0 = \tfrac{3}{2}b\{a - \tfrac{1}{2} + (1-a)^2 \ln(1 + a^{-1})\} \quad (7)$$

where:

$$a = \left(1 + \frac{C^2}{\nu}\right)b \quad (8)$$

$$b = [\mu^{-1} + \nu^{-1}(1 - C)]^{-1}, \quad (9)$$

and σ_0 is the bulk conductivity.

In the case of an infinitely thick film, Equations 7 to 9 take the following limit forms:

$$b_g = \frac{\nu}{1-C}$$

$$a_g = \frac{\nu + C^2}{1-C}$$

$$\sigma_g/\sigma_0 = \frac{3}{2}\frac{\nu}{1-C}\left\{\frac{\nu+C^2}{1-C} - \frac{1}{2} + \left[1 - \left(\frac{\nu+C^2}{1-C}\right)^2\right]\ln\left(1 + \frac{1-C}{\nu+C^2}\right)\right\} \quad (10)$$

in good agreement with previously obtained equations ([1], Equations 17 and 18).

In the case of infinitely thick grains, the limiting forms of Equations 8 and 9 are:

$$a_1 = b_1$$

$$b_1 = \mu.$$

Hence:

$$\sigma_1/\sigma_0 = \tfrac{3}{2}\mu\{\mu - \tfrac{1}{2} + (1 - \mu^2)\ln(1 + \mu^{-1})\}. \quad (11)$$

This is the Cottey equation [8] (Equation 6) as required.

2.2. Linear expression of film conductivity

Assuming that:

$$|b^{-1}| \ll 1, \quad (12)$$

Equation 8 gives: $\quad a \gg 1 \quad$ (12a)

and the function:

$$f(a) = a - \frac{1}{2} + (1 - a^2)\ln\left(1 + \frac{1}{a}\right) \quad (13)$$

takes the form:

$$f(a) \approx \frac{2}{3a}\left(1 - \frac{3}{8a}\right). \quad (14)$$

From Equations 8 and 14, Equation 7 becomes:

$$\sigma_f/\sigma_0 \approx \left(1 + \frac{C^2}{\nu}\right)^{-1}\left[1 - \frac{3}{8a}\right]. \quad (15)$$

According to Equations 8, 9 and 12, the term $[1 - (3/8a)]$ may be replaced by an asymtotic form and Equation 15 becomes:

$$\sigma_f/\sigma_0 \approx \left(1 + \frac{C^2}{\nu}\right)^{-1}\left\{1 - \frac{3}{8}\frac{1-C}{\nu[1 + (C^2/\nu)]}\right\} \times \left\{1 - \frac{3}{8}\frac{1}{[1 + (C^2/\nu) - \frac{3}{8}(1-C)/\nu]\nu}\frac{\nu}{\mu}\right\}. \quad (16)$$

For an infinitely thick film, Equation 16 reduces to:

$$\sigma_g/\sigma_0 \approx \left\{1 - \frac{3}{8}\frac{1-C}{\nu[1 + (C^2/\nu)]}\right\}\left(1 + \frac{C^2}{\nu}\right)^{-1}, \quad (17)$$

where σ_g is the grain-boundary conductivity [1].

An alternative linear form of Equation 16 is then:

$$\sigma_f/\sigma_g \approx 1 - \frac{3}{8}\left[1\Big/\left(1 + \frac{C^2}{\nu} - \frac{3}{8}\frac{1-C}{\nu}\right)\right]\frac{1}{\mu}. \quad (18)$$

An alternative form for the resistivity ρ is

$$\rho_f/\rho_g \approx 1 + \frac{3}{8}\left[1\Big/\left(1 + \frac{C^2}{\nu} - \frac{3}{8}\frac{1-C}{\nu}\right)\right]\frac{1}{\mu}. \quad (19)$$

(Figs. 1 and 2).

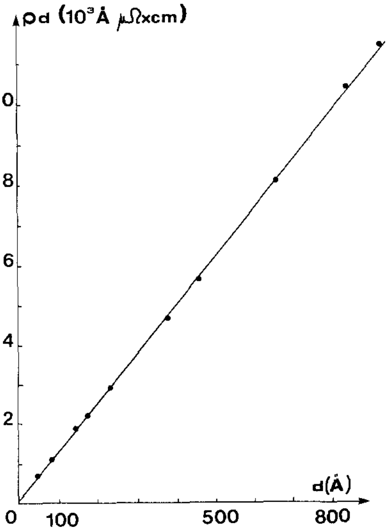

Figure 1 Plots of resistivity thickness, $\rho \times d$, versus thickness, d, of Al annealed r.f. sputtered films.

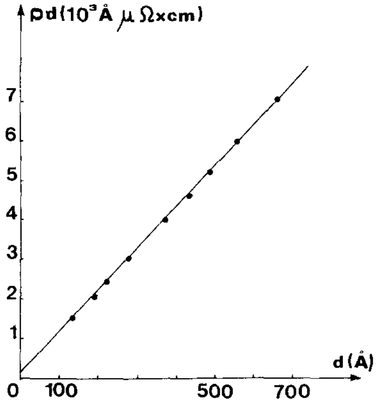

Figure 2 Plots of resistivity thickness, $\rho \times d$: versus thickness, d, of Zn annealed r.f. sputtered films.

These theoretical predictions agree with experiments related to Al and Zn sputtered films, since it has been empirically established [2] that:

$$d \cdot \rho_f = d \cdot \rho_g + E,$$

but it may be noted that Equation 12 may be explicitly written as:

$$\frac{1}{\nu}\left|\left(\frac{\nu}{\mu}+1-C\right)\right| \ll 1 \qquad (20)$$

where $C = 4/\pi$.

From the general theoretical Equation 7, and experimental variations of σ_f [2], the values of ν, t and p have been calculated (Table I). Hence Equation 20 may be replaced by:

$$\left|\frac{\nu}{\mu}+(1-C)\right| < 0.3,$$

i.e.:

$$\frac{\nu}{\mu} < 0.6.$$

Assuming that p and t take similar values, the above condition becomes:

$$\frac{d}{D} > \frac{10}{6}.$$

This condition is always satisfied by continuous sputtered thin films, since the average grain diameter D is usually identified with the first critical thickness [4, 12–14], which corresponds to the thinner continuous film. In addition, in the case of polycrystalline films, the transmission coefficient, t, cannot take values near unity or zero (for $t = 1$ no grain-boundary scattering occurs) and it can be predicted that the grain parameter ν (Equation 5) does not take large values (in agreement with experiments, Table I). Hence it may be concluded that condition 12 does reduce to Equation 20 which is satisfied for continuous films, and Equations 18 and 19 are valid in a large range of thickness, as shown in Table II.

TABLE I

	ν	t	p
Al films	0.52	0.49	0.6
Zn films	1.43	0.42	0.7

2.3. Remarks

Introducing Equation 17 into Equation 16 gives:

$$\sigma_f/\sigma_g \approx 1 - \frac{3}{8}\,\frac{1}{\left(1+\frac{C^2}{\nu}\right)^3(\sigma_g/\sigma_0)}\,\frac{1}{\mu}. \qquad (21)$$

This equation cannot be an effective Fuchs–Sondheimer equation [5], since it cannot derived from the usual asymptotic *F–S* equation [9] by substituting an effective film parameter $\mu_g = \eta \cdot (\sigma_g/\sigma_0)$ for μ. Nevertheless Equation 20 reduces to an effective *F–S* equation for $\nu \gg 1$:

$$\sigma_f/\sigma_g \approx 1 - \frac{3}{8}\,\frac{1}{(\sigma_g/\sigma_0)}\,\frac{1}{\mu} \text{ for } \mu \gg \nu \gg 1. \qquad (22)$$

Equations similar to Equation 22 have been previously presented by several authors [5, 15–18] for an asymptotic description of the conduction properties of polycrystalline films; the above analysis shows that Equations 18 and 19 are more general.

3. Discussion

It must not be neglected that a limitation exists in the validity of the model since the external surface scattering is described by a mean free path; the condition of validity is a large value of the reflection coefficient p [8]; despite that previous numerical studies [5, 15, 18] have shown that such a mean free path may be used for values of p down to 0.2 and it is clear that the situation is the same in this case (Table II).

4. Conclusions

Using the framework of the three-dimensional model of grain-boundary scattering and introducing the Cottey model for the description of scattering at external surfaces, the resistivity of a

TABLE II

	Approx.		Exact [8]	
μ	(Equation 18) σ_F/σ_g	(Equation 19) ρ_F/σ_g	σ_F/σ_g	ρ_F/ρ_g
$\nu = 0.1$				
0.01	−1.056 37	3.056 37	0.376 89	2.653 29
0.02	−0.028 18	2.028 18	0.517 81	1.991 22
0.04	0.485 91	1.514 09	0.660 888	1.513 12
0.1	0.794 36	1.205 64	0.816 797	1.224 29
0.2	0.897 18	1.102 82	0.895 48	1.116 72
0.4	0.948 59	1.051 41	0.943 57	1.059 81
1	0.979 44	1.020 56	0.976 32	1.024 25
2	0.989 72	1.010 28	0.987 96	1.012 19
4	0.994 86	1.005 14	0.993 93	1.006 11
10	0.997 94	1.002 06	0.997 56	1.002 44
$\nu = 0.4$				
0.1	0.293 65	1.706 35	0.601 697	1.661 97
0.2	0.646 83	1.353 17	0.735 750 4	1.399 16
0.4	0.823 41	1.176 59	0.840 382	1.189 94
1	0.923 37	1.070 63		1.079 26
2	0.964 68	1.035 32	0.961 283	1.040 28
4	0.982 34	1.017 66	0.980 094	1.020 31
10	0.992 94	1.007 06	0.991 899	1.008 17
$\nu = 1$				
0.04	−2.442 13	4.442 13	0.293 87	3.402 92
0.08	−0.721 07	2.721 07	0.422 69	2.365 79
0.1	−0.376 85	2.376 85	0.468 41	2.134 89
0.2	0.311 57	1.688 43	0.613 70	1.629 46
0.4	0.655 79	1.344 21	0.745 99	1.340 5
1	0.862 31	1.137 69	0.873 00	1.145 48
2	0.931 16	1.068 84	0.930 44	1.074 76
4	0.965 58	1.034 42	0.963 50	1.037 89
10	0.986 23	1.013 77	0.984 90	1.015 33
$\nu = 4$				
0.1	−1.620 73	3.620 73	0.351 584	2.844 27
0.2	−0.310 36	2.310 36	0.490 403	2.039 14
0.4	0.344 82	1.655 18	0.635 552	1.573 44
1	0.737 93	1.262 07	0.799 126	1.251 37
2	0.868 96	1.131 04	0.884 130 2	1.131 06
4	0.934 48	1.065 52	0.937 072 6	1.067 15
10	0.973 79	1.026 21	0.973 427 2	1.027 30
$\nu = 10$				
0.1	−2.198 68	4.198 68	0.318 07	3.144
0.2	0.599 34	2.599 34	0.451 56	2.214 55
0.4	0.200 33	1.799 67	0.596 63	1.676 08
1	0.680 13	1.319 87	0.768 26	1.301 64
2	0.840 07	1.159 93	0.861 64	1.160 57
4	0.920 03	1.079 97	0.921 532	1.085 15
10	0.968 01	1.031 99	0.963 52	1.037 86

polycrystalline thin metallic films is obtained as a linear function of the reciprocal film thickness. A good fit is obtained with experiments.

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