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Thin polycrystalline metallic-film conductivity under the assumption of isotropic grain-boundary scattering

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The polycrystalline thin-film resistivity and its temperature coefficient of resistivity are calculated from the assumption of isotropic grain-boundary scattering. The proposed simple analytical equations allow separate determination of the transmission coefficient of the grain boundary and the specular reflection coefficient at external surfaces. Good agreement with experiment is found.

1. Introduction

Several attempts have been made recently [1–12] to give a physical description of electrical conductivity in thin metal films. Mayadas and Shatzkes [3] used the theory of quantum mechanics to find a theoretical expression for the film resistivity due to grain boundaries; the size effect in the total film conductivity (including bulk, grain-boundary and external surface scatterings) was expressed [3] in the way previously proposed by Sondheimer [13] for the description of external surface scattering. Since the proposed equations were mathematically complicated and difficult to implement, linearized forms have been studied [14–22] with limited validity ranges.

In the case of annealed films Cottey's analysis [1] for external surface scattering is a convenient tool and gives less sophisticated expressions for the conductivity [9]. The definition of an effective mean free path [7] also allows simple analytical calculations of the conductivity. Moreover, in a large number of cases, the equations of Mayadas and Shatzkes could reduce [6] to Fuchs–Sondheimer equations which are easily manipulated [2]. All these simplified models are convenient tools to describe the film conductivity [6, 7, 21], its temperature coefficient [6, 7] and the strain coefficients [22, 23].

Nevertheless, in the model of Mayadas and Shatzkes the "grain-boundary reflection coefficient" R is defined from a mathematical equation (Equation 6c, in [3]) which is not clearly related to the physical phenomenon of reflection on grain boundaries; moreover, experiments related to annealed, sputtered films gave high values of R [20] which could not easily be interpreted since the contribution of grain-boundary scattering to resistivity is not more than four times that of the bulk scattering [20].

In order to obtain a better representation of the physical phenomena, several statistical models have been proposed [7, 10, 11, 12] which differ from the usual geometrical array of grain boundaries by using cylindrical [10, 24], one-dimensional [7, 11] or three-dimensional [12] arrays. Comparisons with experiment have shown [25] that the three-dimensional model gives a good fit.

The theoretical equations for the conductivity and its temperature coefficient of resistivity (tcr) were easy to use in determining the electrical parameters [25, 26] and could be approximated by linear laws with extended validity ranges [25]. New theoretical expressions for the Hall coefficient [26] in infinitely thick films are similar to those derived [27] from the Fuchs-Sondheimer conduction model [2]; this suggests that the linearized expression for the conductivity could be interpreted in terms of an isotropic additional resistivity due to grain-boundary scattering.

This paper examines theoretical expressions for

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electronic transport derived from the assumption of an isotropic grain boundary mean free path.

2. Theoretical equations

2.1. Expressions for the mean free path

In spherical co-ordinates (r, θ, ϕ) the total electronic mean free path l due to the three types of electronic scattering may be written as [12]

$$l^{-1} = l_0^{-1} + l_i^{-1} + l_c^{-1}, \quad (1)$$

where l_0 is the bulk mean free path and l_i and l_c are related to the grain boundary and the external surface scattering, respectively; they are defined by [11, 12]

$$l_i^{-1} = D^{-1} \ln(1/t)A \quad (2)$$

and

$$l_c^{-1} = d^{-1} \ln(1/p)|\cos\theta|, \quad (3)$$

where D is the average grain diameter, t is the statistically transmission coefficient of any scattering barrier [11, 24], A is the coefficient of the isotropic grain boundary, d is the film thickness and p is the usual [2, 13] electronic specular reflection coefficient at external surfaces.

Introducing Equations 2 and 3 into Equation 1 gives

$$l^{-1} = l_0^{-1}[1 + AD^{-1}l_0 \ln(1/t) + d^{-1}l_0 \ln(1/p) \times |\cos\theta|]. \quad (4)$$

2.2. Theoretical expressions for conductivity and tcr

The conductivity of an infinitely thick film, i.e. the grain boundary conductivity σ_g, may then be derived from the bulk conductivity σ_0 by substituting $l_0^{-1}[1 + AD^{-1}l_0 \ln(1/t)]$ for l_0^{-1} in the well-known expression for the conductivity [2, 13] giving

$$\sigma_g = \sigma_0[1 + AD^{-1}l_0 \ln(1/t)]^{-1}. \quad (5)$$

Similarly the resistivity ratio, ρ_g/ρ_0, is given by

$$\rho_g/\rho_0 = 1 + AD^{-1}l_0 \ln(1/t). \quad (6)$$

Assuming that the thermal linear expansion in D and t is negligible with respect to that of the mean free path, the ratio of the grain boundary tcr, β_g, to the bulk tcr, β_0, is calculated by differentiating Equation 6. This yields

$$\rho_g\beta_g = \rho_0\beta_0, \quad (7)$$

since $\rho_0 l_0$ is independent of temperature [2]. Hence

$$\beta_g/\beta_0 = [1 + AD^{-1}l_0 \ln(1/t)]^{-1}. \quad (8)$$

With the aid of Cottey's procedure [9], the total film conductivity σ_F is given by

$$\sigma_F/\sigma_0 = \frac{3}{4}\int_0^{\pi} \frac{\sin^3\theta}{1 + AD^{-1}l_0 \ln\left(\frac{1}{t}\right) + d^{-1}l_0 \ln\left(\frac{1}{p}\right)|\cos\theta|} \, d\theta.$$

This gives:

$$\sigma_F/\sigma_0 = \frac{3}{2b}[\psi - \tfrac{1}{2} + (1 - \psi^2)\ln(1 + \psi^{-1})] \quad (9)$$

with

$$\psi = [1 + AD^{-1}l_0 \ln(1/t)]b^{-1}, \quad (10)$$

where

$$b = d^{-1}l_0 \ln(1/p). \quad (11)$$

For convenience Equation 9 may be written in the form

$$\sigma_F/\sigma_0 = \frac{1}{b\psi}C(\psi), \quad (12)$$

where

$$C(\psi) = \tfrac{3}{2}\psi[\psi - \tfrac{1}{2} + (1 - \psi^2)\ln(1 + \psi^{-1})] \quad (13)$$

is Cottey's function [1].

Logarithmic differentiation of Equation 13 gives [9]

$$-\beta_F + \beta_0 = -(b\psi)^{-1}\frac{d(b\psi)}{dT} + \frac{D(\psi)}{C(\psi)}\frac{d\psi}{\psi dT}, \quad (14)$$

where

$$D(\psi) = \psi\frac{dC(\psi)}{d\psi} = \tfrac{3}{2}\psi[3\psi - \tfrac{3}{2} + (1 - 3\psi^2)\ln(1 + \psi^{-1})] \quad (15)$$

and

$$\frac{d(b\psi)}{b\psi dT} = \frac{AD^{-1}l_0 \ln(1/t)}{1 + AD^{-1}l_0 \ln(1/t)}\frac{dl_0}{l_0 dT} \quad (16)$$

(from Equation 10), and

$$\frac{d\psi}{\psi dT} = \frac{AD^{-1}l_0 \ln(1/t)}{1 + AD^{-1}l_0 \ln(1/t)}\frac{dl_0}{l_0 dT} - \frac{dl_0}{l_0 dT} \quad (17)$$

(from Equation 11).

Introducing Equations 16 and 17 into Equation 14 and re-ordering gives

$$\beta_F = \beta_0\left(1 - \frac{D(\psi)}{C(\psi)}\right)[1 + AD^{-1}l_0 \ln(1/t)]^{-1}. \quad (18)$$

Equations 9 and 18 can be transformed by introducing Equation 5 and 8, respectively:

$$\sigma_F/\sigma_g = C(\psi) \quad (19)$$

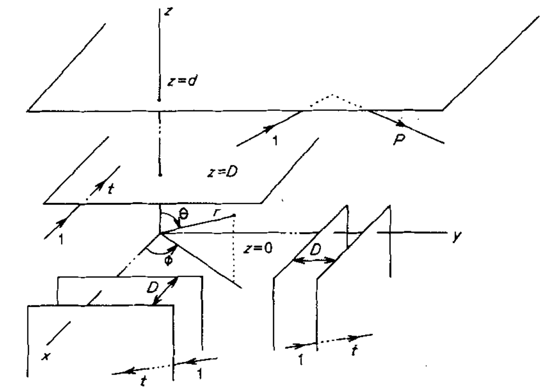

Figure 1 Geometrical model for electronic scattering in polycrystalline thin films.

and

$$\beta_F = \beta_g \left[1 - \frac{D(\psi)}{C(\psi)}\right]. \qquad (20)$$

It may be noted that the effective mean free path conduction model [7] leads to approximate equations for the film resistivity and its tcr which are similar to Equations 19 and 20 ([9], Equations 33a and 33b).

3. Discussion

3.1. Coefficient of isotropic grain-boundary scattering

The value of the coefficient of isotropic grain-boundary scattering A (Equation 2) may be calculated from the general expression for the grain-boundary mean free path calculated in polar co-ordinates (Equation 9 in [12]) (r, θ, ϕ) starting from the framework of a three-dimensional conduction model [12] in which

$$l_i^{-1} = D^{-1} \ln(1/t) [|\cos\phi| \, |\sin\theta| + |\sin\phi| \, |\sin\theta| + |\cos\theta|] \qquad (21)$$

(Fig. 1).

An approximate value for A can be calculated by assuming that the moduli of the sine and cosine terms do not markedly deviate from the average value, i.e.

$$|\cos\phi| = |\sin\phi| = |\sin\theta| = |\cos\theta| \approx 2/\pi. \qquad (22)$$

This gives

$$l_i^{-1} \approx D^{-1} \ln(1/t) \left[\frac{2}{\pi}\left(\frac{4}{\pi} + 1\right)\right]. \qquad (23)$$

Since

$$\frac{2}{\pi}\left(\frac{4}{\pi} + 1\right) \approx 1.45,$$

comparing Equations 2 and 23 gives

$$A \approx 1.45. \qquad (24)$$

3.2. Comparison with the usual model of Mayadas and Shatzkes [3]

A comparison can now be made with the theoretical expressions for the conductivity [3] and tcr [18] of polycrystalline films derived from the Mayadas–Shatzkes model (MS model) [3]. For an easy comparison with numerical data the parameter ψ is rewritten in the form

$$\psi = [1 + Al_0D^{-1} \ln(1/t)][\ln(1/p)]^{-1} d l_0^{-1}. \qquad (25)$$

Introducing the notation of Mayadas and Shatzkes [3]

$$k = dl_0^{-1} \qquad (26)$$

and

$$\alpha = l_0 D^{-1} R(1-R)^{-1}, \qquad (27)$$

where R is the so-called "grain-boundary reflection coefficient", and identifying Equation 5 with the linear asymptotic form ([3], Equation 11) of the grain-boundary resistivity in the MS model, gives the relation

$$1.5R(1-R)^{-1} = A \ln(1/t). \qquad (28)$$

Equation 25 then gives

$$\psi = (1 + 1.5\alpha)[\ln(1/p)]^{-1} k. \qquad (29)$$

Equation 29 allows the calculation of ψ for a given set of values of α and k; numerical data from the MS model are thus reported on the theoretical curves in Figs 2 and 3 corresponding to Equations 19 and 20 for $p = 0.75, 0.5$ and 0.25 and $\alpha = 0.5$, 1 and 2. A deviation of less than 5% is obtained for

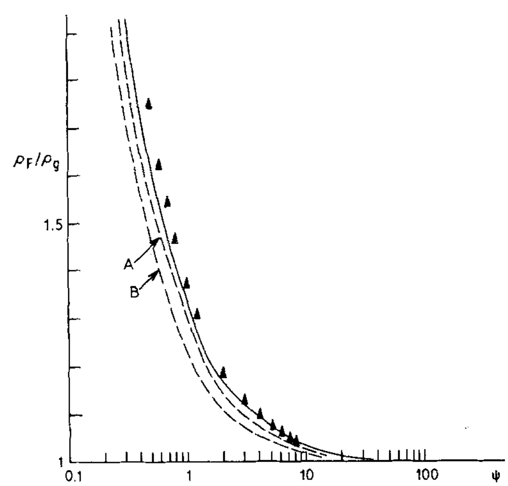

Figure 2 Variation of reduced resistivity ρ_F/ρ_g with the conduction parameter ψ. Full line: theoretical curve from Equation 19 (this cannot be separated from MS curve for $p \geqslant 0.75$); broken lines: MS curves; A, $p = 0.5$; B, $p = 0.25$; Δ: approximate curve (Equation 36).

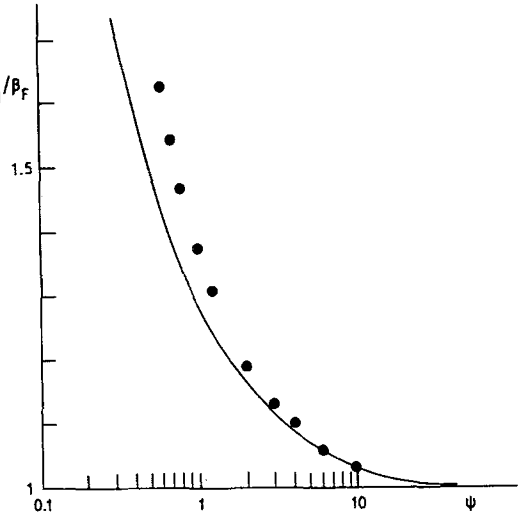

Figure 3 Variation of reciprocal reduced tcr $(\beta_F/\beta_g)^{-1}$ with the conduction parameter ψ. Full line: theoretical curve (Equation 20); •, approximate curve (Equation 37).

$0.5 \leqslant p$ and $0.2 \leqslant \psi$; a deviation of less than 10% is obtained for $0.25 \leqslant p$ and $1 \leqslant \psi$.

3.3. Limiting forms of the theoretical equations

When no grain-boundary scattering occurs, i.e. for $t = 1$ $(\nu \to \infty)$, ψ reduces to $k[\ln(1/p)]^{-1} = \mu$ and Equation 9 becomes

$$\sigma_F/\sigma_0 = \tfrac{3}{2}\mu\,[\mu - \tfrac{1}{2} + (1-\mu^2)\ln(1+\mu^{-1})], \tag{30}$$

which is Cottey's relation [1]. Equation 18 becomes

$$\beta_F/\beta_0 = 1 - \frac{D(\mu)}{C(\mu)}, \tag{31}$$

which is the general expression previously obtained [9].

When ψ becomes infinite, e.g. when the film becomes infinitely thick, the approximate forms of $C(\psi)$ [9] and $D(\psi)$ are:

$$C(\psi) \approx 1 - \frac{3}{8\psi} \tag{32}$$

and

$$D(\psi) \approx \frac{3}{8\psi}. \tag{33}$$

Hence, from Equations 19 and 20

$$(\sigma_F/\sigma_g)_{d\to\infty} \approx 1 - \frac{3}{8\psi} \tag{34}$$

and

$$(\beta_F/\beta_g)_{d\to\infty} \approx 1 - \frac{3}{8\psi} \tag{35}$$

whose alternative forms are

$$\rho_F/\rho_g \approx 1 + \frac{3}{8\psi}, \tag{36}$$

where ρ is the resistivity, and

$$\beta_g/\beta_F \approx 1 + \frac{3}{8\psi}. \tag{37}$$

Deviations of less than 3.2% and 6.6% are obtained for $\psi > 1$ with Equations 36 and 37, respectively.

3.4. Comparison with experiment

It has been recently shown [25] that the variations in resistivity and tcr of thin radio-frequency (r.f.) sputtered Al and Zn films can be described by the following equations

$$\mathrm{d}\rho_F = \mathrm{d}\rho_g + C_1 \tag{38}$$

and

$$\mathrm{d}\beta_F^{-1} = \mathrm{d}\beta_g^{-1} + C_2 \tag{39}$$

with

$$C_1 \approx C_2. \tag{40}$$

These empirical equations agree with Equations 36 and 37 since introducing Equation 25 gives the same asymptotic form for the reduced resistivity and tcr.

$$\rho_F/\rho_g = \beta_g/\beta_F \approx 1 + \tfrac{3}{8}[1 + 1.45\,l_0 D^{-1} \times \ln(1/t)]^{-1} \ln(1/p)\, l_0 d^{-1}. \tag{41}$$

When taking into account Equation 5, Equation 4 may be rewritten in the forms

$$\mathrm{d}\rho_F \approx \mathrm{d}\rho_g + \tfrac{3}{8}\rho_0 l_0 \ln(1/p) \tag{42}$$

and

$$\mathrm{d}\beta_F^{-1} \approx \mathrm{d}\beta_g^{-1} + \tfrac{3}{8}\rho_0 l_0 \ln(1/p). \tag{43}$$

Equations 42 and 43 give a theoretical basis for the validity of Equation 40. Moreover they show that

$$\rho_F\beta_F \approx \rho_g\beta_g.$$

Hence, from Equation 7

$$\rho_F\beta_F \approx \rho_0\beta_0$$

TABLE I Electrical parameters for r.f. sputtered films

R.f. sputtered films	D (nm)	ρ_0 ($\mu\Omega$ cm)	β_0 (10^{-3} K^{-1})	l_0 (nm)	ρ_g ($\mu\Omega$ cm)	β_g (10^{-3} K^{-1})	p	t (from Equation 6)	t (from Equation 8)
Al	11.4	2.65	4.29	31	10.4	1.1	0.595	0.476	0.478
Zn	15.0	5.916	4.19	12	12.2	1.923	0.597	0.4	0.362

TABLE II Values of p and t from resistivity measurements

	p	t
Al films	≈ 0.6	0.49
Zn films	≈ 0.7	0.42

is in good agreement with relations derived [21] from Matthiessen's rule.

The values of the coefficient t (Table I) are determined from the resistivity and tcr ratios according to Equations 6 and 8 by introducing the usual values [2] of the bulk mean free path l_0 and resistivity ρ_0 and the values of the mean grain diameter D which have been previously determined [12]. The values of $\rho_g(\beta_g^{-1})$ are measured from the linear plots [25] of $d\rho_F(d\beta_F^{-1})$ against d (Equations 42 and 43); the values of p (Table I) are deduced from the following equation:

$$\rho_F \approx \rho_g + \tfrac{3}{8}\rho_0 l_0 \ln(1/p)d, \qquad (44)$$

which is derived from Equation 42.

A similar equation related to tcr can be derived from Equation 43 but the experimental inaccuracies [25] in measurements of β_g are more important than those in measurements of ρ and this relation is not retained. For the same reason it is better to use Equation 6 than Equation 8 for calculating t.

The calculated values of p and t are in agreement with those previously obtained (Table II) by a more sophisticated procedure [25] from the framework of three arrays of scatter [12].

Moreover it must be noted that the proposed equations allow separate determinations of p and t whereas the sophisticated procedure requires the best fit to be found when p and t act as parameters. The choice is not always clear: for instance, it has been shown [25] that the value of p in Zn films derived from tcr measurements lies between 0.5 and 0.8.

4. Conclusion

The assumption of isotropic grain-boundary scattering seems an adequate tool for representing the variations in polycrystalline film resistivity and tcr with film thickness and mean grain diameter. Separate determinations of grain-boundary transmission coefficient and specular reflection coefficient are easily performed.

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