Analytical expression for the total electrical conductivity of unannealed and annealed metal films

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Previous studies have shown that the Cottey function constitutes an alternative formulation for the Fuchs–Sondheimer size-effect function, provided that a new parameter is used. This result is used for calculating the effects of scattering at a grain boundary, and a good agreement with the Mayadas–Shatzkes model is found. When background, grain-boundary and external-surface scattering are simultaneously operative, a simple analytical expression for the electrical conductivity of polycrystalline, monocrystalline and columnar metal films can be obtained in the whole experimental domain and may conveniently replace the sophisticated expression of Mayadas and Shatzkes. This expression is similar to that obtained in the framework of the multidimensional models, previously presented. No limitation exists in the value of the electronic specular reflection coefficient, and the theoretical expression is related both to annealed and unannealed films.

1. Introduction

It has been shown in a recent paper [1] that a unique analytical function can be substituted for the Fuchs–Sondheimer size-effect function [2] in thin metal films, whatever the film thickness, d, and the value of the specular electron reflection at film surface, p.

A further development related to the electron transport properties of thin metal films exhibiting grain-boundary scattering is now examined.

2. Theoretical ideas

2.1. The size-effect functions

The Cottey function [1, 3, 4], which is

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

can be used for expressing the size effect in thin metal films in the total range of film thickness and specular reflection coefficient [2], provided that the parameter μ be defined [1] as

$$\mu = \frac{k(1 + p)}{2(1 - p)} \quad (2)$$

where k is the reduced thickness, i.e.

$$k = d\lambda_0^{-1} \quad (3)$$

λ_0 being the electron mean free path in the bulk material. Equations 1 to 3 define the extended Cottey model [1], the e-C model.

Most of the previous theoretical equations [3] have been written with the generalized Cottey parameter μ^* given by

$$\mu^* = k\left(\ln\frac{1}{p}\right)^{-1} \quad (4)$$

and their validity was limited to the range $p > 0.31$ [3, 5]. Linearized equations having a more extended domain of validity have been empirically established [3, 5, 6].

2.2. The grain-boundary effect functions

2.2.1. *The Mayadas–Shatzkes model*

The earliest model for grain-boundary effects was presented by Mayadas and Shatzkes [3, 7]; the grain boundaries were described in terms of dislocation lines perpendicular to the film substrate, but in the case of a very thin film the validity of this description can be criticized [3]. Moreover, the mathematical treatment for calculating the grain-boundary effect was based on several simplifying assumptions [3, 8, 9] (for instance: unidimensional phenomena, multiple layer, no resistivity with regular array of grains). It then did not allow calculations in several dimensions, especially for the Hall coefficient [10].

The electrical conductivity of an infinitely thick film, σ_∞, is given [7] by the relation

$$\sigma_\infty = \sigma_0 f(\alpha)$$

with

$$f(\alpha) = 1 - \tfrac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3\ln(1 + \alpha^{-1}) \quad (5)$$

$$\alpha = \lambda_0 D_g^{-1} R(1 - R)^{-1} \quad (6)$$

where σ_0 is the electrical conductivity of the bulk material, D_g the average grain diameter and R the so-called reflection coefficient at a grain boundary (whose physical interpretation seems questionable [3]).

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2.2.2. The statistical models

The mean free path describing the effect of electron scattering at a grain boundary can be calculated [3, 6, 11] by a procedure similar to that used by Cottey [4] for calculating the effect of scattering at an external film surface. For this purpose a statistical electron transmission coefficient at grain-boundary, t, is defined [3], and the conductivity σ_∞ may be expressed from a Cottey-type function $C(v)$ given by

$$\sigma_\infty = \sigma_0 C(v) \quad (7)$$

where

$$C(v) = \frac{3}{2b_\infty}[a_\infty - \tfrac{1}{2} + (1 - a_\infty^2)\ln(1 + a_\infty^{-1})] \quad (8)$$

with

$$b_\infty = C_2 v^{-1} \quad (9)$$

$$a_\infty = (1 + C^2 v^{-1}) b_\infty^{-1} \quad (10)$$

where

$$C = 4/\pi \quad (11)$$

In the case of polycrystalline films (three-dimensional model [3])

$$C_2 = 1 - C \quad (12)$$

and in the case of monocrystalline and columnar films [6, 11] (bidimensional model [3])

$$C_2 = -C \quad (13)$$

The parameter v was initially defined [3, 11, 12] as

$$v^* = D_g \lambda_0^{-1}\left(\ln \frac{1}{t}\right)^{-1} \quad (14)$$

where the transmission coefficient t plays the same role as the coefficient p in Equation 4. Since the procedure for calculating the associated mean free path is the same, one can predict that the validity range of Equation 8 can be extended to the whole domain provided that v^* is replaced by v:

$$v = D_g \lambda_0^{-1} \frac{1 + t}{2(1 - t)} \quad (15)$$

Since the numerical values of $f(\alpha)$ and $C(v^*)$ roughly coincide [3, 11, 13], if

$$\alpha^{-1} = v^* \quad (16)$$

when the grain-boundary scattering is not very marked, the values of the conductivity of an infinitely thick film of any structure have been calculated (Tables I to III) using either the relation

$$v = (0.9\alpha)^{-1} \quad (17a)$$

TABLE I Compared values of the electrical conductivity of an infinitely thick polycrystalline film in the framework of the Mayadas–Shatzkes model and the three-dimensional model; α is defined by Equation 6 and Equation 17a is used

α	Conductivity	
	M–S model	3-D model
0.010	0.9852	1.0034
0.020	0.9711	0.9779
0.030	0.9574	0.9615
0.040	0.9441	0.9488
0.050	0.9313	0.9361
0.060	0.9189	0.9242
0.070	0.9068	0.9128
0.080	0.8952	0.9015
0.090	0.8838	0.8904
0.100	0.8728	0.8798
0.200	0.7769	0.7853
0.300	0.7012	0.7093
0.400	0.6394	0.6466
0.500	0.5880	0.5942
0.600	0.5444	0.5496
0.700	0.5069	0.5112
0.800	0.4744	0.4779
0.900	0.4458	0.4486
1.000	0.4205	0.4227
2.000	0.2688	0.2681
3.000	0.1977	0.1963
4.000	0.1564	0.1548
5.000	0.1294	0.1278
6.000	0.1103	0.1088
7.000	0.0961	0.0947
8.000	0.0852	0.0839
9.000	0.0765	0.0753
10.000	0.0694	0.0682
20.000	0.0360	0.0353
30.000	0.0243	0.0238
40.000	0.0183	0.0179
50.000	0.0149	0.0144
60.000	0.0124	0.0120
70.000	0.0104	0.0103
80.000	0.0092	0.0090
90.000	0.0077	0.0080
100.000	0.0078	0.0072

TABLE II Compared values of the electrical conductivity of an infinitely thick polycrystalline film in the framework of the Mayadas–Shatzkes model and the three-dimensional model; α is defined by Equation 6 and Equation 17b is used

α	Conductivity	
	M–S model	3-D model
0.010	0.9852	0.9921
0.020	0.9711	0.9657
0.030	0.9574	0.9493
0.040	0.9441	0.9330
0.050	0.9313	0.9178
0.060	0.9189	0.9030
0.070	0.9068	0.8886
0.080	0.8952	0.8746
0.090	0.8838	0.8611
0.100	0.8728	0.8480
0.200	0.7769	0.7362
0.300	0.7012	0.6505
0.400	0.6394	0.5826
0.500	0.5880	0.5276
0.600	0.5444	0.4821
0.700	0.5069	0.4438
0.800	0.4744	0.4111
0.900	0.4458	0.3830
1.000	0.4205	0.3584
2.000	0.2688	0.2184
3.000	0.1977	0.1570
4.000	0.1564	0.1226
5.000	0.1294	0.1005
6.000	0.1103	0.0852
7.000	0.0961	0.0739
8.000	0.0852	0.0653
9.000	0.0765	0.0584
10.000	0.0694	0.0529
20.000	0.0360	0.0271
30.000	0.0243	0.0182
40.000	0.0183	0.0137
50.000	0.0149	0.0110
60.000	0.0124	0.0092
70.000	0.0104	0.0079
80.000	0.0092	0.0069
90.000	0.0077	0.0061
100.000	0.0078	0.0055

TABLE III Compared values of the electrical conductivity of an infinitely thick columnar film in the framework of the Mayadas–Shatzkes model and the bi-dimensional model; α is defined by Equation 6 and Equation 17c is used

α	Conductivity	
	M–S model	2-D model
0.010	0.9852	0.9854
0.020	0.9711	0.9711
0.030	0.9574	0.9574
0.040	0.9441	0.9440
0.050	0.9313	0.9311
0.060	0.9189	0.9185
0.070	0.9068	0.9063
0.080	0.8952	0.8944
0.090	0.8838	0.8828
0.100	0.8728	0.8716
0.200	0.7769	0.7738
0.300	0.7012	0.6966
0.400	0.6394	0.6338
0.500	0.5880	0.5818
0.600	0.5444	0.5379
0.700	0.5069	0.5003
0.800	0.4744	0.4677
0.900	0.4458	0.4392
1.000	0.4205	0.4141
2.000	0.2688	0.2641
3.000	0.1977	0.1942
4.000	0.1564	0.1537
5.000	0.1294	0.1272
6.000	0.1103	0.1085
7.000	0.0961	0.0946
8.000	0.0852	0.0839
9.000	0.0765	0.0753
10.000	0.0694	0.0684
20.000	0.0360	0.0355
30.000	0.0243	0.0240
40.000	0.0183	0.0181
50.000	0.0149	0.0145
60.000	0.0124	0.0121
70.000	0.0104	0.0104
80.000	0.0092	0.0091
90.000	0.0077	0.0081
100.000	0.0078	0.0073

or the relation

$$\nu = (1.18\alpha)^{-1} \quad (17b)$$

both in the framework of the three-dimensional model, or the relation

$$\nu = (1.3\alpha)^{-1} \quad (17c)$$

in the framework of the bi-dimensional model, in order to obtain a good agreement with the Mayadas–Shatzkes equation in the whole range of α, i.e. whatever the grain diameter and the roughness of the grain boundary [6].

Equation 17c is less accurate than Equation 17a, especially at large values of α, but it will be used for expressing the total conductivity (in the next paragraph) because it becomes more accurate in this case.

From Equations 17, the relations between R (Equation 6) and t (Equation 15) can be derived; in the three-dimensional model

$$t = (2 - 2.9R)(2 - 1.1R)^{-1} \quad (18a)$$

and in the bi-dimensional model

$$t = (2 - 3.3R)(2 - 0.7R)^{-1} \quad (18b)$$

Tabulated values are given in Table IV.

TABLE IV Associated values of the grain-boundary coefficient in the Mayadas–Shatzkes mjodel, R, and values of the transmission coefficient t in the bi-dimensional model (2-D) and in the three-dimensional model (3-D) (from Equations 18b and 18a, respectively)

R	Transmission coefficient, t	
	2-D model	3-D model
0.001	0.998	0.998
0.002	0.997	0.997
0.003	0.996	0.996
0.004	0.994	0.995
0.005	0.993	0.994
0.006	0.992	0.992
0.007	0.990	0.991
0.008	0.989	0.990
0.009	0.988	0.989
0.010	0.986	0.988
0.020	0.973	0.976
0.030	0.960	0.964
0.040	0.947	0.952
0.050	0.933	0.939
0.060	0.920	0.927
0.070	0.906	0.914
0.080	0.893	0.902
0.090	0.879	0.889
0.100	0.865	0.876
0.200	0.720	0.742
0.300	0.564	0.596
0.400	0.395	0.435
0.500	0.212	0.257
0.600	−0.012	0.061
0.700	−0.205	−0.158
0.800	−0.444	−0.404
0.900	−0.708	−0.683

2.3. Combined effects of background, grain-boundary and external-surface scatterings

2.3.1. The multidimensional models

Assuming that the reciprocal mean free paths due to any type of scattering may be added for calculating the reciprocal resultant mean free path, a unique expression of the electrical conductivity, σ_f, has been proposed [3, 6, 14] for metal films of any structure (polycrystalline, monocrystalline, columnar) in the general form of a Cottey-type function. Provided that the parameters μ and ν are used (Equations 2 and 15) the preceding paragraphs show that the validity of the equation is extended to the whole domain of thickness and transmission coefficient.

Hence the new equations for the electrical conductivity are

$$\sigma_f = \sigma_0 C(\mu, \nu) \quad (19)$$

with

$$C(\mu, \nu) = \frac{3}{2b}[a - \tfrac{1}{2} + (1 - a^2)\ln(1 + a^{-1})] \quad (20)$$

$$b = \mu^{-1} + C_2\nu^{-1} \quad (21)$$

$$a = b^{-1}(1 + C^2\nu^{-1}) \quad (22)$$

where μ, ν, C_2 and C are defined by Equations 2, 15, 12, 13 and 22, respectively.

2.3.2. The total Mayadas–Shatzkes model

For calculating the total film conductivity, Mayadas and Shatzkes [7] used the Sondheimer procedure [2],

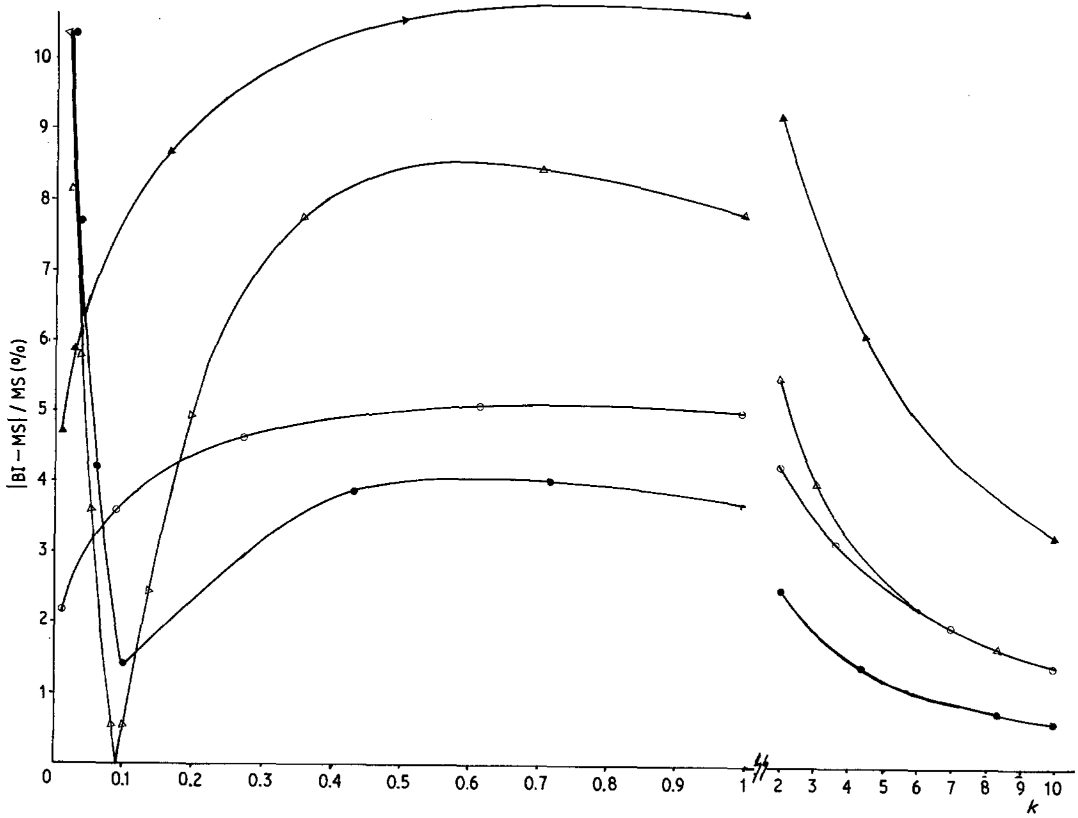

Figure 1 Relative deviation of the theoretical value of the electrical resistivity in the framework of the bidimensional model (BI) (Equations 19 to 22, [13]) from those derived from the Mayadas–Shatzkes model (MS). (▲) $p = 0, \alpha = 0$; (△) $p = 0, \alpha = 1$; (○) $p = 0.25, \alpha = 0$; (●) $p = 0.25, \theta = 1$.

by introducing an effective relaxation time, representing the effect of both background and grain-boundary scattering. The film conductivity [3, 7] is then

$$\sigma_f = \sigma_\infty - \sigma_0 A(k, p, \alpha) = \sigma_0 f_{\text{M-S}}(k, p, \alpha) \quad (23)$$

with

$$A(k, p, \alpha) = \frac{6(1-p)}{\pi k} \int_0^{\pi/2} \mathrm{d}\phi \times \int_1^\infty \mathrm{d}u \frac{\cos^2\phi}{H^2(u, \phi)} (u^{-3} - u^{-5}) \cdots \times \frac{1 - \exp[-kH(u, \phi)]}{1 - p\exp[-kH(u, \phi)]} \quad (24)$$

where

$$H(u, \phi) = 1 + \alpha(\cos\phi)^{-1}(1 - u^{-2})^{-1/2} \quad (25)$$

and u is an integration variable; α is given by Equation 6.

Since the Sondheimer procedure [2] has been used, the remarks related to the Fuchs–Sondheimer conduction model are still valid [1]. The most important feature is that the Fuchs–Sondheimer procedure can be identified with an effective mean free path procedure, since it corresponds either to an exponential distribution in the size-effect mean free path at large thickness or to a Gaussian distribution at low thickness [1]. Hence no theoretical difference exists between the multidimensional models [3, 6] and the total Mayadas–Shatzkes model.

3. Comparison with the Mayadas–Shatzkes model at low values of p

Starting from Equations 17, the numerical values of the total Mayadas–Shatzkes function, $f_{\text{M-S}}(k, p, \alpha)$ (Equation 23), and of the generalized multidimensional functions $C(\mu, \nu)$ (Equation 19) have been calculated for $p < 0.31$ (copies of the detailed numerical data can be obtained from the authors). It is well known that for $p > 0.31$, the numerical values obtained in these models are close together [3, 6].

It appears that the maximal value of the deviation of the multidimensional equations from the Mayadas–Shatzkes equations occurs for the lowest values of k and α and for $p = 0$. The relative deviation does not exceed 11% for $k > 0.01$ in the framework of the three-dimensional model and for $k > 0.02$ in the framework of the bidimensional model (Figs 1 and 2).

One could easily predict this feature since it has recently been shown [1] that the extended Cottey model is an accurate approximate analytical expression to the Fuchs–Sondheimer function.

4. Discussion

The above analysis gives a theoretical basis for the empirical linearized expressions of the reduced resistivity which have recently been proposed [15] in order to extend the limits of the validity [16] of the usual approximate equations [3, 6, 16].

Moreover the above study shows that the approximate equations of the transport parameters expressed in terms of the size parameter, μ, and grain parameter, ν, remain valid at low thickness for unannealed films. Consequently, since most of the electrical parameters can be expressed [3, 6] in terms of the resistivity and/or its temperature coefficient, the effect of annealing on the electrical behaviour of thin metal films can be

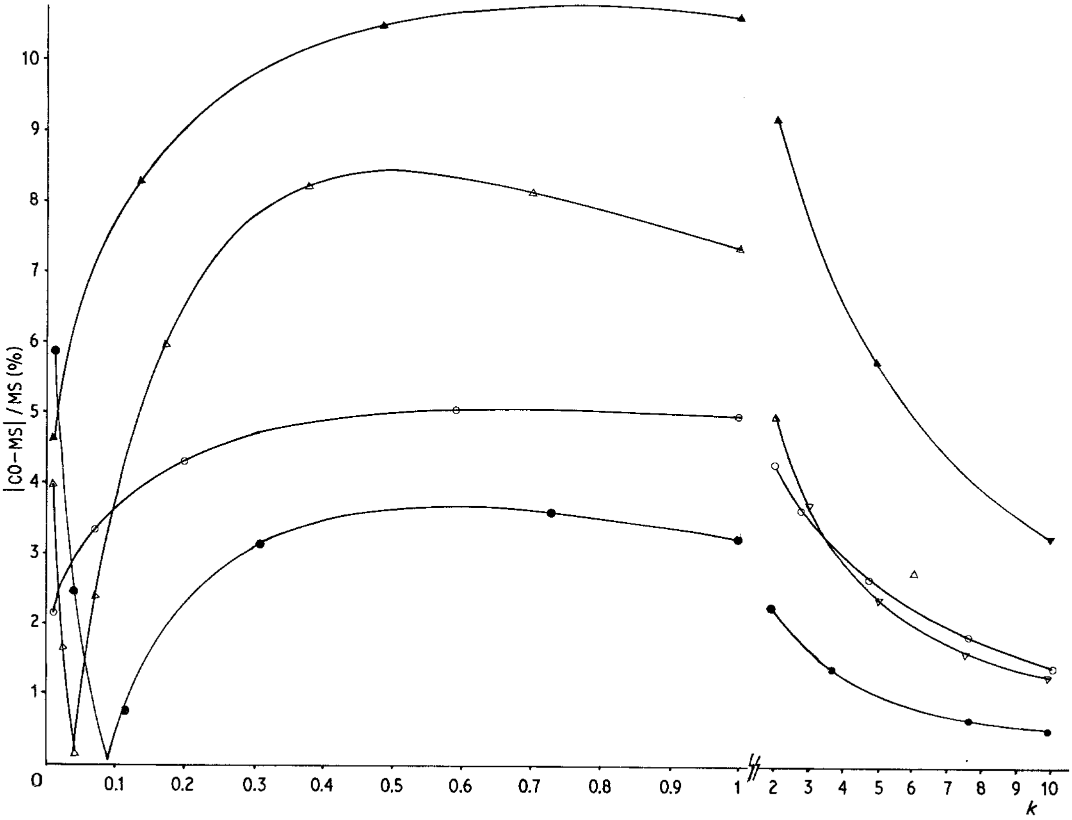

Figure 2 Relative deviation of the theoretical value of the electrical resistivity in the framework of the three-dimensional model (CO) (Equations 19 to 22, [12]) from those derived from the Mayadas–Shatzkes model (MS). (▲) $p = 0, \alpha = 0$; (△) $p = 0, \alpha = 1$; (○) $p = 0.25, \alpha = 0$; (●) $p = 0.25, \alpha = 1$.

studied from Equation 19 and the derived linearized forms [6, 15, 16] for any value of the film thickness.

The fact that the electron specular reflection coefficient could take different values at the top and base film surfaces is not a difficulty, since an average reflection coefficient can be satisfactorily used in this case [17].

More generally, it is clear that the range of validity of the theoretical equations for the electrical resistivity obtained in the frameworks of the multidimensional conduction models [3, 10, 12] can be extended to the whole experimental domain, provided that the new size parameter μ (Equation 2) and the grain parameter ν (Equation 15) are used; for relatively high values of p and t, the more convenient definitions of μ^* (Equation 4) and ν^* (Equation 14) are also valid [3, 6].

The numerical values of Equation 19 can be easily calculated with the aid of a microcomputer (we used a 6502 microcomputer CBM 4062 from Commodore).

5. Conclusion

Whatever the film structure and its electrical state, Equation 19 holds and can be regarded as an alternative algebraic formulation for the sophisticated expression of the total film conductivity obtained in the Mayadas–Shatzkes conduction model of Equation 23.

References

1. C. R. PICHARD, M. BEDDA, C. R. TELLIER, V. I. VATAMANYUK and A. J. TOSSER, *J. Mater. Sci.* **20** (1985) 4185.
2. E. H. SONDHEIMER, *Adv. Phys.* **1** (1952) 1.
3. C. R. TELLIER and A. J. TOSSER, "Thin Film Science and Technology 2", (Elsevier, Amsterdam, 1982) Ch. 1.
4. A. A. COTTEY, *Thin Solid Films* **1** (1967/68) 297.
5. C. R. TELLIER, C. R. PICHARD and A. J. TOSSER, *J. Mater. Sci. Lett.* **1** (1982) 271.
6. C. R. PICHARD, Thèse de Doctorat-ès-Sciences, Université de Nancy-I (1985).
7. A. F. MAYADAS and M. SHATZKES, *Phys. Rev. B* **1** (1970) 1382.
8. C. R. TELLIER and A. J. TOSSER, *Le Vide, Couches Minces* **189** (1977) 121.
9. C. R. PICHARD, C. R. TELLIER and A. J. TOSSER, *ibid.* **408** (1981) 619.
10. C. R. TELLIER and A. J. TOSSER, "Thin Film Science and Technology 2" (Elsevier, Amsterdam, 1982) Ch. 2.
11. C. R. TELLIER, C. R. PICHARD, V. I. VATAMANYUK and A. J. TOSSER, *J. Mater. Sci. Lett.* **2** (1983) 579.
12. C. R. PICHARD, C. R. TELLIER and A. J. TOSSER, *Thin Solid Films* **62** (1979) 189.
13. C. R. TELLIER and A. J. TOSSER, *ibid.* **70** (1980) 225.
14. C. R. PICHARD, V. I. VATAMANYUK, A. KHALID-NACIRI, C. R. TELLIER and A. J. TOSSER, *J. Mater. Sci. Lett.* **3** (1984) 447.
15. M. BEDDA, Thesis, Université de Nancy-I (1985).
16. V. I. VATAMANYUK, A. J. TOSSER, C. R. PICHARD and C. R. TELLIER, *J. Mater. Sci.* **19** (1984) 4138.
17. L. GUENDOUZ, C. R. TELLIER, A. J. TOSSER and C. R. PICHARD, *J. Mater. Sci. Lett.* **3** (1984) 377.

Received 18 March
and accepted 17 June 1985