JOURNAL OF MATERIALS SCIENCE 26 (1991) 1421–1430

A Soffer–Cottey model for the galvanomagnetic properties of thin polycrystalline metal films

C. R. TELLIER
Laboratoire de Chronométrie, Electronique et Piézoélectricité, Ecole Nationale supérieure de mécanique et des microtechniques, Route de Gray, La Bouloie, 25030 Besançon Cedex, France

Theoretical equations for the galvanomagnetic parameters of thin polycrystalline metal films are derived within the frameworks of the Soffer–Cottey model for electron scattering at rough surfaces and of the three-dimensional model for electron scattering at grain boundaries. Influences of the surface roughness and of the strength of the transverse magnetic field are investigated, showing the absence of oscillations in the transport parameters with increasing magnetic field even for very thin films with rough surfaces. In the limits of small reduced thicknesses and weak magnetic fields, analytical expressions for the Hall coefficient and for the electrical conductivity are proposed in order to provide an easier experimental determination of the roughness and grain parameters.

1. Introduction

In the past few years, interest in the theoretical study of variations in the Hall coefficient or in the magnetoresistance of thin metal films with the film thickness has been revived [1–5]. Let us recall that in the classical Fuchs–Sondheimer model (FS model) [6, 7] for the size effects in thin films, a constant specularity parameter p was introduced to describe the scattering of carriers at the external surfaces. Then, recently some theoretical work [3, 8, 9] was devoted to more or less sophisticated extensions of the FS model. These extensions, which deal with the influence of the angle, θ, of the incident wave vector and of r.m.s. height of the roughness, r, at film surfaces are based on the Soffer model [10] of an angularly-dependent specularity. Among these studies, Preist and Sambles [3] extended the FS model and observed oscillations in electrical conductivity as a function of the magnetic field in the case of films submitted to an electric field $E(E_x, E_y, 0)$ and a transverse magnetic field $H(0, 0, H)$. However, Tellier [4, 5] combined the Cottey formalism [11] and the Soffer model to express the total relaxation time associated with the simultaneous background and external surface scattering, and obtained a monotonic increase in the film conductivity with the magnetic field strength, even for very strong magnetic fields. But the oscillations in the film conductivity sometimes observed in experiments [11, 12] have also been interpreted in terms of deviations from the free electron model [13, 14] and attributed to quantified momentum orbitals. However, the problem up to now has not been completely elucidated and there is no reason to make the Cottey formalism more questionable than the FS formalism.

Moreover, a great deal of experimental work [15–25] on the morphology of as-deposited as well as of thermally-annealed thin metal films has given evidence for films having high defect densities. Point defects and dislocations are currently observed [15, 22, 26, 27] but in addition films are frequently found to be grained [15–25]. Grained films can exhibit a columnar structure; however, examination of the morphology of films by scanning electron microscopy, transmission electron microscopy and X-ray diffractometry, also reveals fine-grained structures [15–21]. Thus the contribution of the grain boundary scattering to size effects in the transport parameters needs also to be taken into account when attempting to interpret data on the galvanomagnetic properties of films in terms of an angularly dependent surface scattering. Here it must be pointed out that except for the theoretical work recently carried out by Warkusz [9], which is essentially concerned with thermal effects in polycrystalline films and in which some partial results on the influence of the surface roughness are derived at the light of the FS model, there are to our knowledge no complete theoretical studies devoted to the surface roughness dependence of the galvanomagnetic properties of polycrystalline films.

Thus the purpose of this paper is to use the Cottey formalism to derive the formulations for the galvanomagnetic properties of thin polycrystalline metal films, which include the contribution of the r.m.s. surface roughness and of the angle of incidence to the total relaxation time of the carriers. The influence of the surface roughness parameter on the magnetoresistance and on the Hall coefficient is systematically investigated. Emphasis is placed on the magnetic field dependence of the galvanomagnetic properties. Some attempts are made to extract the contribution of the grain-boundary scattering to the total size effect.

0022-2461/91 $03.00 + .12

2. Theoretical treatment

2.1. Expressing the distribution function of carriers

In the case of polycrystalline films we have to take into account three different scattering processes going on at the same time:

1. The *background scattering*, which is described by a relaxation time τ_0. The background relaxation time includes the elastic scattering of carriers by point defects, and for a sake of simplicity it is supposed to be independent of the film thickness, d.

2. The *carrier scattering at the external surface*, for which a relaxation time τ_s can be defined. In a combined SC model the relaxation time τ_s is given by [4, 28]

$$\tau_s = d\left[v_F \cdot \ln\left(\frac{1}{p}\right)|\cos\theta|\right]^{-1} \qquad (1)$$

where v_F is the velocity of carriers at the Fermi surface. The specularity parameter p is that calculated by Soffer [10] in the framework of classical models for electromagnetic waves scattering at rough surfaces [29].

$$p = \exp\left[-\left(\frac{4\pi r}{\lambda_c}\cos\theta\right)^2\right] \qquad (2)$$

where λ_c is the Fermi wavelength of conduction electrons. Equation 1 can be rewritten in the form

$$\tau_s = \tau_0 A \cos^2\theta|\cos\theta| \qquad (3)$$

involving the size parameter A, given by

$$A = k^{-1}(4\pi r/\lambda_c)^2 \qquad (4)$$

k being the reduced thickness, i.e. the ratio of the film thickness, d, to the background mean free path, λ_0.

3. The *carrier scattering at grain boundaries*, where we choose for the relaxation time τ_g the expression derived several years ago by Tellier and Tosser [1, 30] who assumed that the grain boundaries can be represented by three arrays of planar boundaries with rough surfaces respectively perpendicular to the x, y and z axes and that from a statistical point of view only a fraction t of the carriers are specularly transmitted through any potential associated with the grain boundaries. To avoid having to deal with a multitude of unknown parameters it is usual [1] to assume crudely that for fine-grained films the grains are cubic. In these conditions the relaxation time τ_g related to the simultaneous background and grain-boundary scattering processes is given by [30]

$$\tau_g = \tau_0[1 + C^2\nu^{-1} + (1 - C)\nu^{-1}|\cos\theta|]^{-1} \qquad (5)$$

with $C = 4/\pi$.

In the special case of cubic grains, the grain parameter ν is related to the average grain diameter D by means of the formula

$$\nu = D\lambda_0^{-1}[\ln(1/t)]^{-1} \qquad (6)$$

If the scattering events occur independently of each other, then Matthiessen's rule is fulfilled and we find the equation for the total relaxation time, $\tau(\theta)$, as

$$\tau(\theta) = \tau_0[\mathscr{F}(\theta)]^{-1} \qquad (7)$$

where the angularly dependent function $\mathscr{F}(\theta)$ is given by

$$\mathscr{F}(\theta) = 1 + \nu^{-1}C^2 + (1 - C)\nu^{-1}|\cos\theta| + A|\cos\theta|\cos^2\theta \qquad (8)$$

We now proceed to calculate the distribution function $f = f^0 + f^1$ for a geometry related to a metal film placed in an electric field $\boldsymbol{E}(E_x, E_y, 0)$ and a transverse magnetic field $\boldsymbol{H}(0, 0, H)$. The deviation f^1 from the equilibrium function f^0 caused by the external electrical and magnetic fields is the solution of the equation [1, 31]

$$\frac{f^1}{\tau(\theta)} - \frac{eH}{m}\left(v_y\frac{\partial f^1}{\partial v_x} - v_x\frac{\partial f^1}{\partial v_y}\right) = \frac{e}{m}\left(E_x\frac{\partial f^0}{\partial v_x} + E_y\frac{\partial f^0}{\partial v_y}\right) \qquad (9)$$

where v_x and v_y are components of the free electron velocity $\boldsymbol{v}$ (absolute value v). e is the absolute charge of an electron and m is the effective electron mass.

For the purpose of determining the solution of Equation 9 we follow a treatment previously proposed by Sondheimer [7]. Let us represent f^1 by means of two functions C_1 and C_2 which do not depend explicitly on v_x and v_y:

$$f^1 = (v_x C_1 + v_y C_2)\frac{\partial f^0}{v} \qquad (10)$$

Then let us define the complex quantities

$$g = C_1 - jC_2 \qquad (11a)$$

$$F = E_x - jE_y \qquad (11b)$$

in such a way that the substitution of Equation 10 into Equation 11 yields a pair of simultaneous equations which with the use of Equations 11 can be rewritten as a compact equation

$$g[\tau(\theta)]^{-1} + jgvr_B^{-1} = e(mv)^{-1}F \qquad (12)$$

where r_B is the radius of the Larmor orbit of an electron moving in a magnetic field of magnitude H:

$$r_B = mv(eH)^{-1} \qquad (13)$$

We readily find the solution of Equation 12

$$g = \frac{e}{mv}\left((E_x - jE_y)\left[\tau_0^{-1}\mathscr{F}(\theta) - j\frac{v\tau_0}{r_B}\right] \Big/ \left\{\tau_0^{-2}\left[[\mathscr{F}(\theta)]^2 + \left(\frac{v\tau_0}{r_B}\right)^2\right]\right\}\right) \qquad (14)$$

Here for convenience we use α to denote the field parameter

$$\alpha = \lambda_0/r_B \qquad (15)$$

By turning to Equation 11a we finally get the equations for the functions C_1 and C_2 as

$$C_1 = \frac{e\tau_0}{mv}\left(\frac{\mathscr{F}(\theta)E_x - \alpha E_y}{[\mathscr{F}(\theta)]^2 + \alpha^2}\right) \qquad (16)$$

$$C_2 = \frac{e\tau_0}{mv}\left(\frac{\mathscr{F}(\theta)E_y + \alpha E_x}{[\mathscr{F}(\theta)]^2 + \alpha^2}\right) \tag{17}$$

2.2. Expressing the galvanomagnetic parameters of polycrystalline films

We have now to calculate the current density (J_x, J_y, 0). This requires the evaluation of integrals of the form [1, 7]

$$J_x = -2e\left(\frac{m}{h}\right)^3 \iiint C_1 v_x^2 \frac{\partial f^0}{\partial v} \mathrm{d}^3 v \tag{18}$$

$$J_y = -2e\left(\frac{m}{h}\right)^3 \iiint C_2 v_y^2 \frac{\partial f^0}{\partial v} \mathrm{d}^3 v \tag{19}$$

For this purpose, as a first step we introduce spherical coordinates in v-space with $v_z = v\cos\theta$. After carrying the integrations over the variables v and φ, Equations 18 and 19 become

$$J_x = \frac{3}{4}\sigma_0(E_x I_1 - \alpha E_y I_2) \tag{20}$$

$$J_y = \frac{3}{4}\sigma_0(E_y I_1 - \alpha E_x I_2) \tag{21}$$

where I_1 and I_2 denote integrals over the variable θ:

$$I_1 = \int_0^\pi \frac{\sin^3\theta \mathscr{F}(\theta)}{[\mathscr{F}(\theta)]^2 + \alpha^2]} d\theta \tag{22}$$

$$I_2 = \int_0^\pi \frac{\sin^3\theta}{[\mathscr{F}(\theta)]^2 + \alpha^2} d\theta \tag{23}$$

and σ_0 is the background conductivity.

If in a second step we transform the integration variable from θ to $u = \cos\theta$, the alternative expression for the current density is found to be given by

$$J_x = \frac{3}{2}\sigma_0(E_x \mathscr{A} - \alpha E_y \mathscr{B}) \tag{24}$$

$$J_y = \frac{3}{2}\sigma_0(E_y \mathscr{A} + E_x \mathscr{B}) \tag{25}$$

with

$$\mathscr{A} = \int_0^1 \frac{[1 + C^2\nu^{-1} + (1 - C)\nu^{-1}u + Au^3](1 - u^2)}{[1 + C^2\nu^{-1} + (1 - C)\nu^{-1}u + Au^3]^2 + \alpha^2} \mathrm{d}u \tag{26}$$

$$\mathscr{B} = \int_0^1 \frac{1 - u^2}{[1 + C^2\nu^{-1} + (1 - C)\nu^{-1}u + Au^3]^2 + \alpha^2} \mathrm{d}u \tag{27}$$

To express the galvanomagnetic parameters for the geometry under consideration we start from the theoretical formulae for the film conductivity:

$$\left[\sigma_f = \frac{J_x}{E_x}\right]_{J_y = 0} \tag{28}$$

for the magnetoresistance M_f:

$$M_f = \frac{\Delta\rho_f}{\rho_{f,0}} = \frac{\rho_f(H) - \rho_f(0)}{\rho_f(0)} \tag{29}$$

and for the Hall coefficient:

$$\left[R_{Hf} = \frac{E_y}{J_x H}\right]_{J_y = 0} \tag{30}$$

Setting $J_y = 0$ in Equation 25 and eliminating the components E_x and E_y of the electric field in Equations 28 and 30, we get

$$\frac{\sigma_0}{\sigma_f} = \frac{3}{2}\left(\frac{\mathscr{A}}{\mathscr{A}^2 + \alpha^2\mathscr{B}^2}\right) \tag{31}$$

$$\frac{R_{Hf}}{R_{HO}} = \frac{2}{3}\left(\frac{\mathscr{B}}{\mathscr{A}^2 + \alpha^2\mathscr{B}^2}\right) \tag{32}$$

Equation 32 is obtained by taking into account the fact that for bulk metals the Hall coefficient is related to the number n of free electrons by

$$R_{HO} = -1/ne \tag{33}$$

The film magnetoresistance is easily found to be

$$M_f = \mathscr{A}_0\mathscr{A}(\mathscr{A}^2 + \alpha^2\mathscr{B}^2)^{-1} - 1 \tag{34}$$

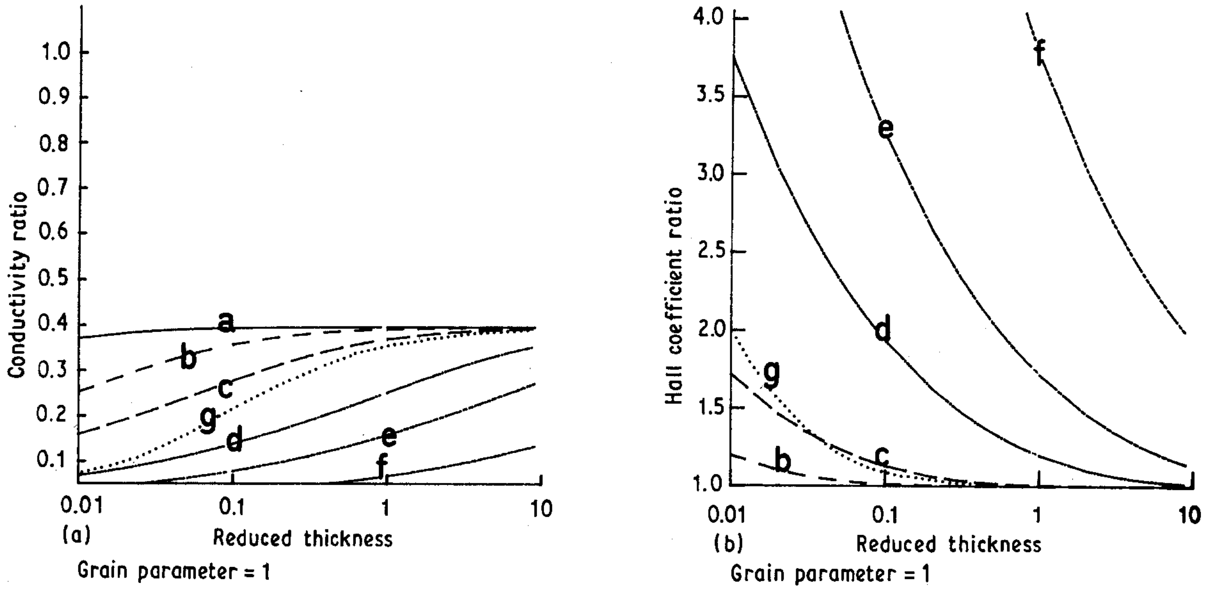
where $\mathscr{A}_0$ denotes the reduced form of the integral $\mathscr{A}$ obtained by setting $\alpha = 0$.

3. Discussion and conclusion

3.1. Presentation of theoretical results

Theoretical values of the galvanomagnetic parameters have been evaluated using standard computational techniques. In order to give a rapid overview of the influence of the surface roughness on the transport parameters, results of such computations are shown in Fig. 1a, b and c for a moderate contribution to the resistivity by grain boundaries ($\nu = 1$). On these figures we have also displayed when appropriate the curves corresponding to a constant value of 0.5 for the specularity parameter p as drawn from the three-dimensional model of grain boundaries [32, 33]. It appears reasonable to assume that the main contribution to the current is due to electrons impinging on external surfaces at $\theta = \pi/2$; then, turning to the expression for p (Equation 2) in the combined Soffer–Cottey (SC) and three-dimensional (TD) model (SC–TD model), we readily find that the corresponding value for the reduced roughness is close to 0.1. At first sight the curves resulting solely from three-dimensional type calculations [32, 33] are in close agreement with the present curves. But after a precise examination it is clear that there are several interesting differences:

(i) Comparing the predictions of the TD model with those of the SC–TD model it appears that the effect of the angular dependence of p is to decrease the overall size effect in the conductivity. A similar behaviour for the film conductivity was previously reported by Tellier [28], neglecting the grain boundary scattering process.



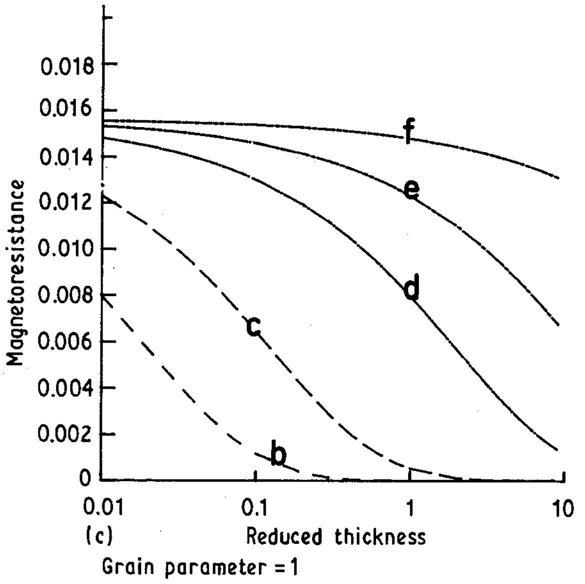


Figure 1 The influence of the surface roughness on the various transport parameters of a polycrystalline metal film with $\nu = 1$. (a) σ_f/σ_0 against k for $\alpha = 1$. Curves a, b, c, d, e and f are for r/λ_c = 0.01, 0.04, 0.1, 0.4, 1 and 4 respectively. Curve g is the Cottey curve for $p = 0.5$. (b) R_{Hf}/R_{H0} against k for $\alpha = 1$. Curves b, c, d, e and f are for $r/\lambda_c = 0.04, 0.1, 0.4, 1$ and 4. Curve g is the Cottey curve for $p = 0.5$.

(ii) In the absence of a contribution of grain boundaries to the transport properties the SC model also gives a decrease in the overall size effect in the Hall coefficient [5]. Moreover, for k lying in the range 0.01 to 10 the Hall coefficient as evaluated in the framework of the TD model [34] always remains higher than the Hall coefficient as given by the SC theory [5]. Here again the SC–TD model gives an overall size effect which is apparently less than in the TD theory (Fig. 1b). However, the TD and SC–TD curves cross each other for values of k in the vicinity of 0.07. Then at higher k values we can strongly conclude that the effect of the angular and roughness dependence of p is to decrease the apparent influence of the electron scattering process at external surfaces.

However, although the TD and SC–TD models exhibit some differences the SC–TD model satisfies some essential physical requirements whatever the strength of magnetic field:

(i) From Table I, which incorporates the influence of the magnetic field strength on the galvanomagnetic parameter through the parameter α, we observe that progressively transforming a smooth surface to a rough surface causes a monotonic increase in R_{Hf}/R_{H0} and a monotonic decrease in σ_f/σ_0. Such a behaviour has been previously depicted by Tellier [4, 5] in the limit of weak and strong magnetic fields when developing the SC model.

(ii) As expected, the size effect in galvanomagnetic parameters vanishes for large reduced thickness. Here the predicted behaviour agrees with all the conduction models such as the Sondheimer model, the SC model and so on.

The influence of the electron scattering at grain boundaries on the transport parameter may be seen through Table II. We observe that as the grain boundaries act as more and more efficient scatterers (i.e. for decreasing ν), all the galvanomagnetic parameters, M_f, σ_f/σ_0 and R_{Hf}/R_{H0} take smaller and smaller values. In particular, one may see that the grain-boundary scattering process partially alters the influence of the surface roughness on the transverse magnetoresistance and on the Hall coefficient. One cannot then reasonably expect to observe experimentally a measurable magnetoresistance effect in fine-grained films ($\nu < 0.4$), even for very thin films ($k \leqslant 0.01$). We note that as the grain-boundary scattering becomes less and less efficient the values for the galvanomagnetic parameters tend to the SC-type values.

Now we concentrate our attention on the theoretical variations in the galvanomagnetic parameters with the field parameter $\beta = k\alpha$. At this point let us recall that some authors [3] developing calculations in the framework of the FS model obtained oscillations in the conductivity ratio for $\beta > 1$ with a first

TABLE I The galvanomagnetic parameters for various values of the surface roughness parameter, r/λ_c and the field strength assuming $k = 0.01$ and moderately efficient scattering at grain boundaries ($\nu = 0.4$)

$\alpha = 0.9$				
r/λ_c	0.01	0.04	0.1	0.4
M_f	2.4×10^{-5}	0.00026	0.00250	0.0033
σ_f/σ_0	0.200409	0.15099	0.09953	0.04381
R_{Hf}/R_{HO}	1.00079	1.11042	1.48795	3.09547
$\alpha = 9$				
r/λ_c	0.01	0.04	0.1	0.4
M_f	0.06500	0.06853	0.15201	0.20716
σ_f/σ_0	0.20028	0.14135	0.08661	0.03641
R_{Hf}/R_{HO}	1.00021	1.06477	1.03701	2.76388
$\alpha = 90$				
r/λ_c	0.01	0.04	0.1	0.4
M_f	0.00088	0.19585	0.06810	2.23558
σ_f/σ_0	0.20024	0.12630	0.04245	0.013584
R_{Hf}/R_{HO}	1.00000	1.00227	1.05778	1.06954

TABLE II The galvanomagnetic parameters for various values of the grain parameter ν and the field strength, assuming $k = 0.01$ and moderately rough surfaces ($r/\lambda_c = 0.04$)

$\alpha = 0.9$					
ν	0.2	0.4	0.8	2	4
M_f	0.00027	0.00131	0.00465	0.01495	0.02562
σ_f/σ_0	0.09260	0.15084	0.22623	0.33245	0.39786
R_{Hf}/R_{HO}	1.05593	1.10963	1.17784	1.26631	1.31576
$\alpha = 9$					
ν	0.2	0.4	0.8	2	4
M_f	0.01945	0.06853	0.16376	0.33561	0.45811
σ_f/σ_0	0.09086	0.141352	0.19530	0.25264	0.27985
R_{Hf}/R_{HO}	1.04204	1.06477	1.08103	1.09255	1.09667
$\alpha = 90$					
ν	0.2	0.4	0.8	2	4
M_f	0.08464	0.19585	0.36868	0.64828	0.83862
σ_f/σ_0	0.08540	0.12630	0.16606	0.20471	0.22193
R_{Hf}/R_{HO}	1.00209	1.00227	1.00237	1.00242	1.00244

minimum in σ_f/σ_0 occurring for β close to unity. The relative magnitudes of the oscillations in conductivity are much more for rough surfaces than for smooth surfaces. The oscillations tend to disappear for small r/λ_c ($r/\lambda_c < 0.04$). Increasing the reduced thickness from $k = 0.01$ to $k = 0.5$ causes a marked decrease in the oscillations. Moreover, variations in R_{Hf}/R_{HO} with β also show oscillations at high β ($\beta > 1$) centred about $R_{Hf}/R_{HO} = 1$ [3].

Going back to the present model, Fig. 2 illustrates the Hall coefficient result for $\nu = 1$. It should be pointed out that the galvanomagnetic parameter, which is less sensitive to the grain-boundary scattering process, is just the Hall coefficient. Thus a general conclusion for the Hall coefficient behaviour can easily be drawn from the results displayed in Fig. 2. In the present model we never observe oscillations in the Hall coefficient, even for very thin films with rough surfaces ($r/\lambda_c > 1$). In this limit of strong magnetic fields we can consider Fig. 3 as representative of the general behaviour for the variations in the conductivity ratio σ_f/σ_0 with the field parameter β. From Fig. 3 we infer that even for very thin films ($k < 0.01$) the conductivity ratio decreases monotonically to give a plateau for a limiting value β_L of the field parameter. Increasing the film thickness ($k = 0.1$) results in a shift of the plateau which moves toward the higher field parameter region. Clearly the SC–TD model yields no oscillatory pattern in the polycrystalline film conductivity as a function of increasing magnetic field. As the field parameter region investigated here corresponds to that studied by Preist and Sambles [3], we disagree with these authors about the prediction of a decaying oscillating behaviour in the field dependence of the conductivity. Moreover, if we look at plots of the transverse magnetoresistance M_f against the field parameter β, for different values of the film thickness (Fig. 4) we also observe the absence of an oscillatory

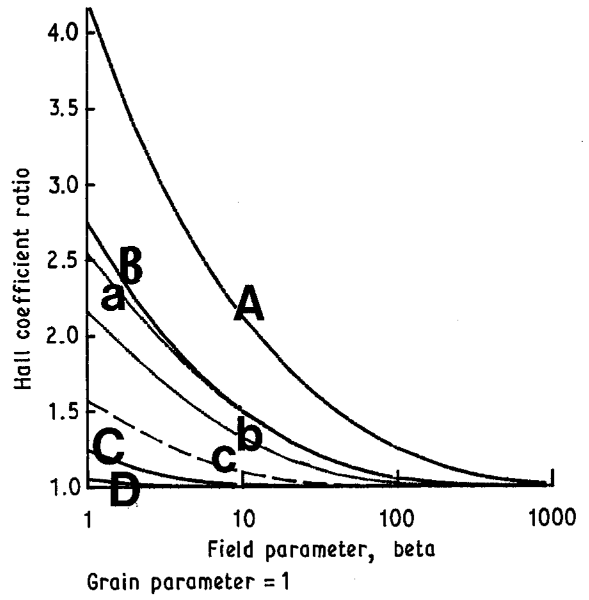


Figure 2 The field parameter, β, variation of the Hall coefficient ratio, R_{Hf}/R_{H0}, for thin polycrystalline films with $\nu = 1$. Curves A, B, C and D are for $k = 0.01$ and for $r/\lambda_c = 2$, 1, 0.2 and 0.1, respectively. Curves a, b and c are for $k = 0.1$ and for $r/\lambda_c = 1$, 075 and 0.4, respectively.

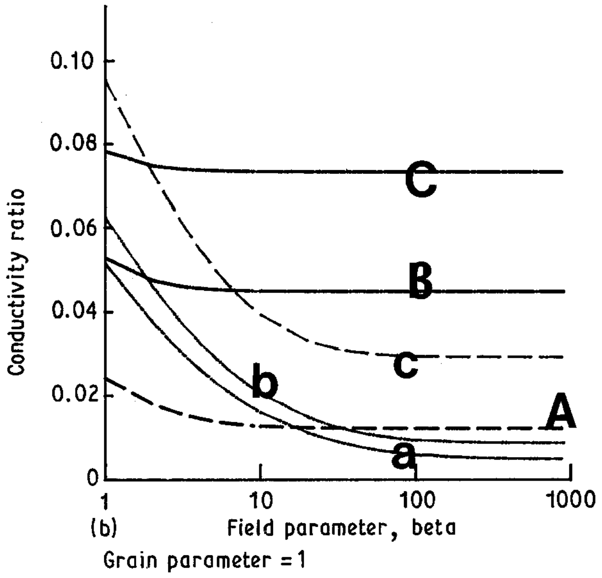


Figure 3 The field parameter, β, variation of the conductivity ratio, σ_f/σ_0 for thin polycrystalline films with $\nu = 1$. Curves A, B, C are for $k = 0.01$ and for $r/\lambda_c = 0.2$, 0.1 and 0.075, respectively. Curves a, b, c are for $k = 0.1$ and for $r/\lambda_c = 1$, 0.75 and 0.4, respectively.

pattern. Effectively the transverse magnetoresistance shows a steady increase with increasing field strength, followed in the strong field region by a quasi-independence of the field parameter β.

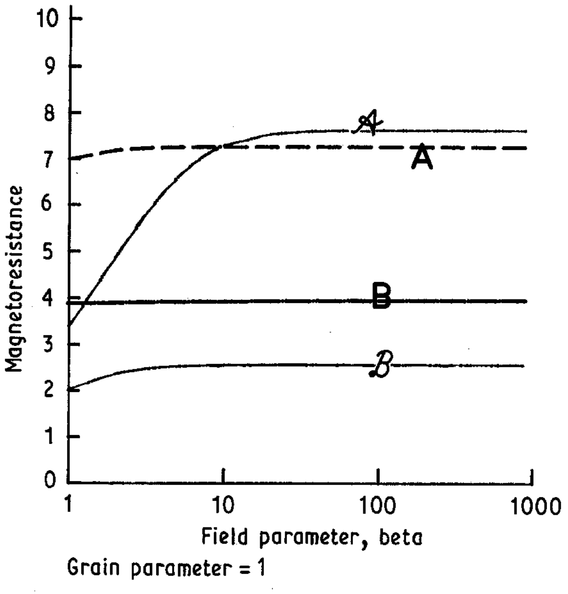


Figure 4 The field parameter, β, variation of the magnetoresistance M_f for thin polycrystalline films with $\nu = 1$. Curves A and B are for $k = 0.01$ and for $r/\lambda_c = 0.2$ and 0.1, respectively. Curves $\mathscr{A}$ and $\mathscr{B}$ are for $k = 0.001$ and for $r/\lambda_c = 0.06$ and 0.04, respectively.

3.2. Analytical expressions for the galvanomagnetic parameters

Using Equations 31, 32 and 34, which do not express analytically the galvanomagnetic parameters, to determine experimental values for the reduced roughness r/λ_c and the grain parameter ν leads to practical difficulties in detecting easily the exact contribution of r/λ_c and ν to the galvanomagnetic properties. Direct measurements of r/λ_c and ν require effectively a separation of the contributions to the galvanomagnetic properties of the surface roughness, of the film thickness and of the grain parameter. Owing to the fact that the Hall coefficient exhibits large size effects exclusively when we are concerned with very thin films ($k < 0.1$) with rough surfaces ($r/\lambda_c > 0.06$), it seems interesting to concentrate our attention on measurements made on thin films at low temperatures for which the size effects in R_{Hf} and M_f are not usually too limited in magnitude. Moreover, only weak magnetic fields are easily available in experiments. The purpose of this section is therefore to derive simple equations for the Hall coefficient and the conductivity in the limiting cases of small reduced thickness and weak magnetic field, in order to perform an easy determination of the roughness parameter from experimental data.

Useful information about the nature of the dependence of the Hall coefficient on the reduced thickness can be obtained from Fig. 5, which represents plots of $\ln(R_{Hf}/R_{H0})$ against $\ln k$, with r/λ_c and ν acting as parameters. We obtain straight lines with a slope of about $-1/6$. Repeating this procedure for plots of the variations of $\ln(R_{Hf}/R_{H0})$ against $\ln(r/\lambda_c)$ (insert to Fig. 5) and against $\ln \nu$ (Fig. 6) also gives linear variations from which we can deduce values of about 2/3 and 2/11 for the respective slopes. The effect of the external surfaces and grain-boundary scatterings on the Hall coefficient ratio is then easily estimated to be of the form

$$\ln(R_{Hf}/R_{H0}) \approx \tfrac{2}{3}\ln(r/\lambda_c) - \tfrac{1}{3}\ln k + \tfrac{2}{11}\ln \nu + \mathscr{C}_R$$

$$\alpha \ll 1, k \ll 1 \qquad (35)$$

$\mathscr{C}_R$ is a constant which one can adjust to minimize in the large ν and r/λ_c ranges any discrepancies between the exact and approximate values of the Hall coefficient ratio. Table III provides further information

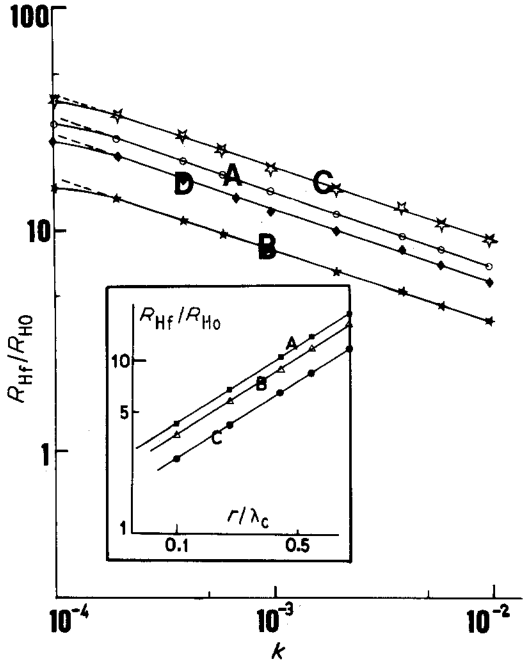


Figure 5 Plots of $\ln(R_{Hf}/R_{H0})$ against $\ln k$ with ν and r/λ_c acting as parameters. Curves A, B, C, D are for $\nu = 1$ and $r/\lambda_c = 1$; $\nu = 1$ and $r/\lambda_c = 0.4$; $\nu = 4$ and $r/\lambda_c = 1$; $\nu = 0.4$ and $r/\lambda_c = 1$, respectively. In the inset are shown plots of $\ln(R_{Hf}/R_{H0})$ against $\ln(r/\lambda_c)$ for $k = 0.001$. Curves A, B, C are for $\nu = 10$, 2 and 0.6, respectively.

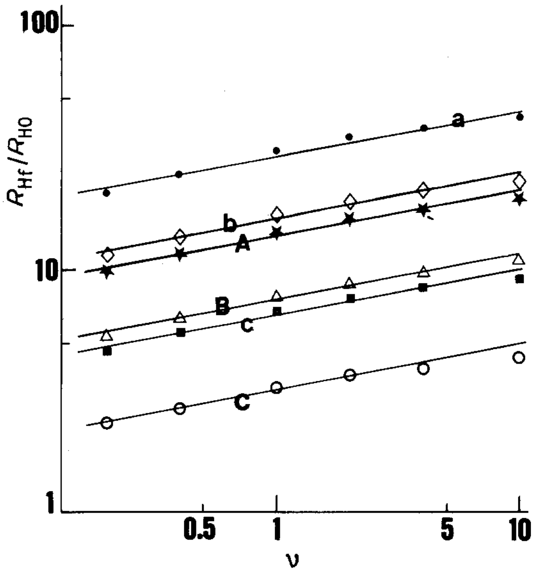


Figure 6 Plots of $\ln(R_{Hf}/R_{H0})$ against $\ln \nu$. Curves a, b, c are for $k = 0.0001$ and for $r/\lambda_c = 1$, 0.4 and 0.1, respectively. Curves A, B, C are for $k = 0.001$ and for $r/\lambda_c = 1$, 0.4 and 0.1, respectively.

about the ranges of applicability of the approximate equation [35]. We observe that the approximate form of R_{Hf}/R_{H0} accurately represents the exact form down to $k \approx 0.01$ until the grain parameter lies in the range 0.6 to 6. Increasing the reduced roughness results in an extension of the k range of applicability of the approximate equation. As an example, for $\nu = 0.6$ the deviation between the exact and approximate values of R_{Hf}/R_{H0} as given by Equations 32 and 35, respectively, remains less than 10% for $k \leq 0.3$ as r/λ_c increases from 0.1 to 1.

To discuss the influence of the reduced surface roughness on the conductivity of polycrystalline films we need to deal with an approximate equation which makes additive the various contributions to the resistivity. Attempts to plot $\ln(\sigma_f/\sigma_0)$ against $\ln k$ and $\ln(\sigma_f/\sigma_0)$ against $\ln(r/\lambda_c)$ yield straight lines with slopes respectively equal to about 1/3 (Fig. 7) and − 2/3 (inset to Fig. 7).

Then, despite the opposite sign, we obtain similar values for the magnitudes of the slopes relating to corresponding linear plots (see insets to Figs 5 and 7, for example). This similarity is destroyed when we turn our attention to the dependence of the conductivity ratio on the grain parameter. By plotting $\ln(\sigma_f/\sigma_0)$ against ν, k and r/λ_c acting as parameters, we find (Fig. 8) a more complex situation: all the points do not lie on a single straight line. In reality the conductivity date lie crudely on two interesting straight lines corresponding to a large ν ($\nu > 2$) and a small ν ($\nu < 1$) region. The procedure yields a line of lower slope for the large ν region than for the small ν region. Then the function which approximates the exact variations in the conductivity with k, r/λ_c and ν may be

$$\ln(\sigma_f/\sigma_0) \approx \tfrac{1}{3}\ln k - \tfrac{2}{3}\ln(r/\lambda_c) + \tfrac{1}{6}\ln \nu + \mathscr{C}_c, \qquad \alpha \ll 1, k \ll 1 \tag{36}$$

in the limit of large ν, whereas an estimate of σ_f/σ_0 may be given by

$$\ln(\sigma_f/\sigma_0) \approx \tfrac{1}{3}\ln k - \tfrac{2}{3}\ln(r/\lambda_c) + \tfrac{6}{11}\ln \nu + \mathscr{C}_c, \qquad \alpha \ll 1, k \ll 1 \tag{37}$$

in the small ν region.

To make the comparison between the exact and approximate values of σ_f/σ_0 more significant, values of the conductivity as evaluated from the exact Equation 31 and from the respective Equations 36 and 37 are listed in Tables IV and V. It can be seen that when the contribution of grain boundaries to the resistivity remains weak, the discrepancies between the exact and the approximate values do not exceed 8% for $k \leq 0.03$. But as soon as we are concerned with fine-grained films with rough surfaces, Table V reveals small deviations (typically less than 6%) between exact and approximate values in a relatively large range of k value ($k \leq 0.1$). Numerical values displayed in Table V show also that the k range of applicability of Equation 37 depends markedly on the value of the reduced roughness; the smoother are the external surfaces, the smaller the k range of applicability of Equation 37 becomes. At this point it should be noticed that the first and second terms of the approximate Equations 36 and 37 resemble those previously proposed by Tellier [35] in the limit of large A to estimate the conductivity of metal films with rough surfaces in which the grain-boundary scattering is inoperative. Effectively Tellier showed that in the combined Soffer–Cottey model the approximate equation

$$\ln\frac{\sigma_f}{\sigma_0} \approx \frac{1}{3}\ln k - \frac{2}{3}\ln\frac{r}{\lambda_c} + \ln\frac{\pi}{3^{1/2}} - \frac{2}{3}\ln 4\pi \tag{38}$$

holds for large A.

TABLE III The Hall coefficient ratio R_{Hf}/R_{H0} in the limit of small magnetic fields, as evaluated from the exact Equation 32 and the approximate Equation 35. The value omitted from this table corresponds to a physically unreasonable value

k	$\nu = 0.6$		$\nu = 2$		$\nu = 6$	
	Eq. 32	Eq. 35	Eq. 32	Eq. 35	Eq. 32	Eq. 35
$r/\lambda_c = 0.1$						
0.0001	6.076	6.087	7.663	7.576	8.604	9.252
0.0003	4.281	4.220	5.372	5.253	6.020	6.415
0.001	2.960	2.825	3.677	3.516	4.105	4.294
0.003	2.162	1.959	2.644	2.438	2.935	2.977
0.01	1.596	1.311	1.900	1.632	2.086	1.993
0.03	1.281	–	1.469	1.131	1.587	1.382
$r/\lambda_c = 1$						
0.0001	27.706	28.253	35.149	35.167	39.551	42.942
0.0003	19.233	19.590	24.390	24.384	27.441	29.775
0.001	12.909	13.114	16.356	16.323	18.395	19.932
0.003	8.991	9.093	11.375	11.317	12.786	13.820
0.01	6.076	6.087	7.663	7.576	8.604	9.252
0.03	4.281	4.220	5.372	5.253	6.020	6.415

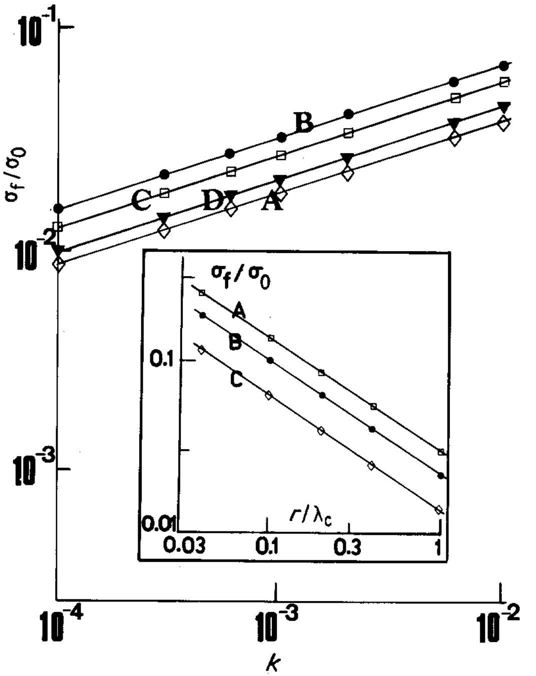


Figure 7 Plots of $\ln(\sigma_f/\sigma_0)$ against $\ln k$, with ν and r/λ_c acting as parameters. Curves A, B, C, D are for $\nu = 1$ and $r/\lambda_c = 1$; $\nu = 1$ and $r/\lambda_c = 0.4$; $\nu = 4$ and $r/\lambda_c = 1$; $\nu = 0.4$ and $r/\lambda_c = 1$, respectively. In the inset are shown plots of $\ln(\sigma_f/\sigma_0)$ against $\ln(r/\lambda_c)$ for $k = 0.001$. Curves A, B, C are for $\nu = 10$, 2, and 0.6, respectively.

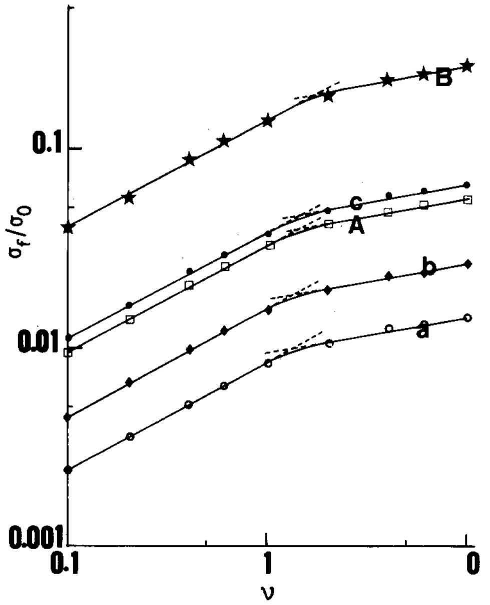


Figure 8 Plots of $\ln(\sigma_f/\sigma_0)$ against $\ln \nu$. Curves a, b, c are for $k = 0.0001$ and for $r/\lambda_c = 1$, 0.4 and 0.1, respectively. Curves A and B are for $k = 0.006$ and $r/\lambda_c = 1$ and 0.1, respectively.

Finally we may observe that we can also optimize values for the constants $\mathscr{C}_R$ and $\mathscr{C}_c$. However, such research constitutes at this stage a speculative refinement because firstly the curves describing the exact and approximate variations in R_{Hf}/R_{H0} with the film thickness cross each other, and secondly the main difficulties for the film conductivity arise when we try to track the influence of the grain parameter by means of a single equation.

3.3. Conclusion

Combining the Soffer–Cottey model and the three-dimensional model of grain boundaries, we have presented results for the electrical conductivity, the Hall coefficient and the transverse magnetoresistance. The results differ from those derived in the framework of the classical Sonheimer model. The major fact is that even in the regime of small thickness and strong magnetic fields, all the galvanomagnetic parameters vary monotonically to reach limiting values with increasing β. Clearly any oscillatory phenomenon in the galvanomagnetic parameter arises from increasing magnetic field. At this point it should be remarked that the classical Sondheimer theory was developed under the assumption of quasi-free electrons so that the Fermi surface is spherical. However, it is now well established [14] that the appearance of oscillations in the conductivity and in the Hall coefficient as the field is varied is connected to types of Fermi surface which

TABLE IV The thickness variation of the conductivity ratio σ_f/σ_0 as evaluated from the exact Equation 31 and the approximate Equation 36: the limits of small magnetic fields and weak contributions to the resistivity by grain boundaries

k	$\nu = 2$		$\nu = 4$		$\nu = 10$	
	Eq. 31	Eq. 36	Eq. 31	Eq. 36	Eq. 31	Eq. 36
$r/\lambda_c = 1$						
0.0001	0.01048	0.01013	0.01241	0.01137	0.01409	0.01324
0.0003	0.01511	0.01461	0.01789	0.01639	0.02031	0.01910
0.001	0.02256	0.02182	0.02671	0.02449	0.03032	0.02853
0.003	0.03249	0.03147	0.03848	0.03532	0.04368	0.04115
0.01	0.04839	0.04701	0.05732	0.05277	0.06508	0.06147
0.03	0.06941	0.06780	0.08227	0.07610	0.09343	0.08866
$r/\lambda_c = 0.1$						
0.0001	0.04839	0.04701	0.05732	0.05277	0.06508	0.06147
0.0003	0.06947	0.06780	0.08227	0.076101	0.09343	0.088657
0.001	0.10251	0.10128	0.12164	0.11368	0.13825	0.13244
0.003	0.14497	0.14607	0.17239	0.16395	0.19620	0.19101
0.01	0.20821	0.21819	0.24862	0.24491	0.28372	0.28533
0.03	0.28184	0.31469	0.33871	0.35323	0.38819	0.41151

TABLE V The thickness variation of the conductivity ratio σ_f/σ_0 as evaluated from the exact Equation 31 and the approximate Equation 37: the limits of small magnetic fields and very fine-grained films

k	$r/\lambda_c = 1$				$r/\lambda_c = 0.1$			
	$\nu = 0.6$		$\nu = 0.2$		$\nu = 0.6$		$\nu = 0.2$	
	Eq. 31	Eq. 37	Eq. 31	Eq. 37	Eq. 31	Eq. 37	Eq. 31	Eq. 37
0.001	0.00651	0.00667	0.00357	0.00348	0.02997	0.03097	0.01636	0.01618
0.003	0.00938	0.00962	0.00515	0.00503	0.04290	0.04466	0.02331	0.02333
0.001	0.01400	0.01437	0.00767	0.00751	0.06305	0.06672	0.03392	0.03486
0.003	0.02015	0.02073	0.01103	0.01083	0.08845	0.09622	0.04686	0.05027
0.01	0.02997	0.03097	0.01636	0.01618	0.1251	0.14374	0.06441	0.07510
0.03	0.04290	0.04466	0.02331	0.02333	0.1655	0.20731	0.08184	0.10831
0.1	0.06304	0.06667	0.03392	0.03486	0.2114	0.30968	0.09858	0.16180

ensure an extremal and singular value for the derivative $(\partial\mathscr{A}/\partial k_H)$ where $\mathscr{A}$ is the area of the Fermi surface and k_H is the component of the wave vector $\boldsymbol{k}$ along $\boldsymbol{H}$. These singularities arise when we are concerned with a truncated Fermi surface, an inflection point or an elliptic limiting point on the Fermi surface. Clearly spherical Fermi surfaces do not contain such singularities, and there is no reason to attribute the predicted oscillations to properties of localized regions of the Fermi surface. This is the reason why the present calculations, which are of course for a free-electron metal, do not predict an oscillatory behaviour. Moreover, in a recent study of the transverse magnetoresistance of thin potassium films, Gridin *et al.* [36] observed no oscillations in the transverse magnetoresistance as the field strength is increased. According to Sondheimer's prediction the potassium films are expected to exhibit at least one, two and seven oscillations in the range of field strength investigated by these authors, and some attempts have been made without success to explain the absence of oscillations in terms of the usual arguments such as a large Hall field or the presence of open orbits due to preferred orientation.

The experimental determination of parameters related to the morphology and the geometrical surface states of thin films remains an open question. The possibility of fitting data implies at least a separation of the parameters related to the different scattering processes. For this reason approximate equations for the Hall coefficient ratio and the conductivity ratio are proposed in the low-field regime. These equations concern essentially thin films ($k < 0.1$) for which the problem of separating the different contributions to the Hall effect turns out to be more tractable. The proposed equations seem convenient to follow, for example, the changes in the surface roughness induced by successive thermal annealing, provided that the intermediate and final measurements of R_{Hf} and σ_f were performed at relatively low temperatures. It should be noticed that annealing causes, at the lower annealing temperatures, a mechanical reordering of the top surface of the film [1, 18, 37–41] which is followed at higher annealing temperatures by an increase in the average grain diameter [1, 15, 24, 38, 39, 42–44]. Care must then be taken in the interpretation of experiments. In particular, simultaneous measurements of the size effect on various parameters must be made (here at least on σ_f and R_{Hf} (together with a systematic investigation of the changes in grain size with annealing treatment. Since the transmission coefficient t certainly remains slightly affected by annealing, one can reasonably attribute changes in ν to variations in D. Moreover, variations in σ_0 and λ_0 on annealing may be also interpreted in terms of a reduction of the concentration of other frozen-in structural defects such as point defects or lines of dislocations [15, 27, 44, 45]. Provided the annealing behaviour of the various defects are carefully examined, the proposed approximate equations appear to be convenient tools to estimate the variation in the surface roughness caused by the thermal ageing.

References

1. C. R. TELLIER and A. J. TOSSER, "Size Effect in Thin Films" (Elsevier, Amsterdam, 1982).
2. G. GOVINDARAJ and V. DEVANATHAN, *Phys. Rev. B* **32** (1985) 2628.
3. T. W. PREIST and J. R. SAMBLES, *J. Phys. F., Met. Phys.* **16** (1986) 2119.
4. C. R. TELLIER, *J. Mater. Sci.* **22** (1987) 2906.
5. *Idem., ibid.* **23** (1988) 1464.
6. K. FUCHS, *Proc. Cambridge Phil. Soc.* **34** (1938) 100.
7. E. H. SONDHEIMER, *Phys. Rev.* **80** (1950) 401.
8. J. R. SAMBLES and K. C. ELSOM, *J. Phys. D* **15** (1982) 1459.
9. F. WARKUSZ, *Thin Solid Films* **148** (1987) 343.
10. S. SOFFER, *J. Appl. Phys.* **38** (1967) 1710.
11. A. A. COTTEY, *Thin Solid Films* **1** (1967/68) 297.
12. K. FORSVOLL and I. HOLWECH, *Phil. Mag.* **9** (1964) 435.
13. M. INEOU, Y. TAMAKJ and H. YAGI, *J. Appl. Phys.* **45** (1974) 1562.
14. C. M. HURD, "The Hall Effect in Metals and Alloys" (Plenum, London, 1982).
15. A. GANGULEE, *J. Appl. Phys.* **43** (1972) 3943.
16. U. ADMON and A. BAR-OR, D. TREVES, *ibid.* **44** (1973) 2300.
17. R. H. CORNELY and T. A. ALI, *ibid.* **49** (1978) 4094.
18. T. T. SHENG, R. B. MARCUS, F. ALEXANDER and W. A. REED, *Thin Solid Films* **14** (1972) 289.
19. P. A. B. TOOMBS and P. BENNETT, *J. Appl. Phys.* **39** (1968) 2948.
20. V. DAMODARA DAS and J. C. MOHANTY, *ibid.* **54** (1983) 977.
21. D. DAYAL, P. RUDOLF and P. WISSMANN, *Thin Solid Films* **79** (1981) 193.
22. S. SEN, R. K. NANDI and S. P. SEN GUPTA, *ibid.* **48** (1978) 1.
23. M. MURAKANI, *ibid.* **59** (1979) 105.
24. G. J. VAN GURP, *ibid.* **38** (1976) 295.
25. H. T. G. NILSSON, B. ANDERSSON and S. E. KARLSSON, *ibid.* **63** (1979) 87.
26. S. D. MUKHERJEE, in "Reliability and Degradation", edited by M. J. Howes and D. V. Morgan (Wiley, Chichester, 1981) Ch. 1.
27. R. SURI, A. P. THAKOOR and K. L. CHOPRA, *J. Appl. Phys.* **46** (1975) 2574.
28. C. R. TELLIER, *J. Mater. Sci. Lett.* **3** (1984) 464.
29. P. BECKMANN and A. SPIZZICHINO, "The Scattering of Electromagnetic Waves from Rough Surfaces" (Pergamon, London, 1963).
30. C. R. TELLIER, C. R. PICHARD and A. J. TOSSER, *Thin Solid Films* **61** (1979) 349.
31. J. M. ZIMAN, "Electrons and Phonons" (Oxford University Press, London, 1962).
32. C. R. PICHARD, A. J. TOSSER and C. R. TELLIER, *J. Mater. Sci.* **16** (1981) 451.
33. C. R. PICHARD, C. R. TELLIER and A. J. TOSSER, *Thin Solid Films* **62** (1979) 189.
34. C. R. TELLIER, M. RABEL and A. J. TOSSER, *J. Phys. F., Met. Phys.* **8** (1978) 2357.
35. C. R. TELLIER, *J. Mater. Sci.* **20** (1985) 4514.
36. V. V. GRIDIN, W. R. DATARS and Y. B. NING, *J. Phys. Con. Mater.* **1** (1989) 713.
37. R. E. HUMMEL and H. F. GEIER, *Thin Solid Films* **25** (1977) 335.
38. C. R. TELLIER and A. J. TOSSER, *ibid.* **43** (1977) 261.
39. C. R. TELLIER, *Vacuum* **28** (1978) 321.
40. A. J. TOSSER, C. R. TELLIER and J. LAUNEY, *ibid.* **27** (1977) 261.
41. J. M. CHAUVINEAU and C. PARISET, *Surf. Sci.* **36** (1973) 1955.
42. P. WISSMANN, *Thin Solid Films* **6** (1970) R67.
43. J. M. HERAS and E. E. MOLA, *ibid.* **35** (1976) 75.
44. G. J. VAN GURP, *J. Appl. Phys.* **46** (1975) 1922.
45. J. M. HERAS and E. V. ALBANO, *Thin Solid Films* **106** (1983) 275.

Received 11 January
and accepted 28 March 1990