JOURNAL OF MATERIALS SCIENCE 23 (1988) 1464–1473

Theoretical size effects in galvanomagnetic properties of thin metal films in a Soffer–Cottey model: interpretation of experiments

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

In defining an effective relaxation time which depends on the root mean square (r.m.s.) surface roughness and on the angle of incidence of electrons, theoretical results on the electrical conductivity, the magnetoresistance and the Hall coefficient in thin metal films subjected to a transverse magnetic field have been extensively presented. Except for the magnetoresistance, a decrease in the overall size effect is observed in transport parameters with respect to the predictions of classical theories based on the Fuchs–Sondheimer or the Cottey models. The size effect in the product resistivity × temperature coefficient of resistivity is found to be correlated with that in the normalized Hall coefficient. Tentative attempts to fit previously published data to framework of the combined Soffer–Cottey model are undertaken. As a result, difficulties in choosing reasonable values for the bulk parameter in the limit of very small reduced thicknesses are outlined. In the regime of relatively large reduced thicknesses, emphasis is placed on the requirement of the simultaneous measurements of various transport parameters on the same metal films and of a systematic control of the surface texture and the morphology of films to provide a meaningful interpretation of experimental data.

1. Introduction

It is well known that the transport properties of thin metal films can be significantly modified with respect to bulk properties by the additional scattering of the charge carriers at the film surfaces [1]. These size effects have generally been accounted for [1–3] in terms of the theory developed by Sondheimer [4] who imporved some earlier calculations derived by Fuchs [5] in 1938. The most important feature of the Fuchs–Sondheimer theory is that a constant specular reflection coefficient, P, is defined as the fraction of the carriers that are scattered specularly at the film surfaces. Later some authors extended the Fuchs–Sondheimer theory to include the cases where the angle of incidence, θ, at the film surfaces [6–8] or the root mean square (r.m.s.) surface roughness r [9] changes the specularity of the carrier scattering. Finally, Soffer [10] proposed a theory where both the r.m.s. surface roughness and the angle of incidence influence the specularity parameter. When the correlation length along the film surface is taken to be zero the Soffer model gives the following expression for the specularity parameter

$$p = \exp\left[-\cos^2\theta\left(4\pi\frac{r}{\lambda_c}\right)^2\right] \qquad (1)$$

where λ_c is the wavelength associated with the carrier.

In the past few years new theoretical results on the film conductivity were worked out by Sambles and Elsom [11, 12] who followed the framework of the Fuchs–Sondheimer theory and used Equation 1 for the specularity parameter. Unfortunately, their equations must be treated numerically and thus it remains difficult to compare rapidly experimental data and theoretical predictions. Recently, Tellier [13] proposed an alternative treatment of the size effect in the electrical conductivity of thin metal films in which the Cottey model [14] is combined with the Soffer model (SC model). The SC model which expresses the reduced film conductivity analytically in terms of the reduced r.m.s. surface roughness, r/λ_c, and of the reduced thickness, k, also gives approximate equations for the conductivity of metal films which are convenient tools for an easy determination of the roughness parameter from experimental data [15]. Tellier [16] also calculated the effects of the r.m.s. surface roughness and of the angle of incidence on the electrical conductivity and on the Hall coefficient of thin metal films subjected to a transverse magnetic field. As in this case, the final equations are somewhat complicated in such a way that a numerical integration becomes necessary; simple analytical equations have been proposed [16] in the weak- and strong-field limits.

The purpose of this paper is to give a complete overview of the influence of the surface roughness on the transport parameters without limitation on the strength of the magnetic field. The main features revealed by the theoretical model are intensively discussed. Emphasis is made on the size effects in

0022-2461/88 $03.00 + .12

transverse magnetoresistance. Finally, some experimental works are reinterpreted in the light of these new theoretical results.

2. Theory

2.1. Theoretical equations

Let us recall that in the framework of the SC model [13, 16] and for the geometry illustrated in Fig. 1, the relaxation time $\tau(\theta, r)$ which describes the simultaneous background scattering and the electron scattering at the external surfaces is given by

$$\tau(\theta, r) = \tau_0(1 + A \cos^2 \theta |\cos \theta|)^{-1} \tag{2}$$

where τ_0 is the background relaxation time. The influence of the reduced r.m.s. roughness, r/λ_c, and of the ratio, k, of the film thickness, d, to the background mean free path λ_0 is seen through the parameter A.

$$A = \frac{1}{k}\left(\frac{4\pi r}{\lambda_c}\right)^2 \tag{3}$$

The transport Boltzmann equation for a thin metal film placed in an electric field $\boldsymbol{E}(E_x, E_y, 0)$ and a transverse magnetic field $\boldsymbol{H}(0, 0, H)$ (Fig. 1) was previously solved following a classical procedure proposed earlier by Sondheimer [17]. After algebraic manipulations the electric current densities, J, in the x direction and the y direction are finally found to be expressed as

$$J_x = \tfrac{3}{2}\sigma_0\{E_x\mathscr{A} - \alpha E_y\mathscr{B}\} \tag{4}$$

$$J_y = \tfrac{3}{2}\sigma_0\{E_y\mathscr{A} + \alpha E_x\mathscr{B}\} \tag{5}$$

with

$$\mathscr{A} = \int_0^1 \frac{(1 + Au^3)(1 - u^2)}{(1 + Au^3)^2 + \alpha^2}\,\mathrm{d}u \tag{6}$$

$$\mathscr{B} = \int_0^1 \frac{1 - u^2}{(1 + Au^3)^2 + \alpha^2}\,\mathrm{d}u \tag{7}$$

where u is an integration variable, and σ_0 is the background conductivity. α is the usual field parameter [17] defined by

$$\alpha = \lambda_0/r_B \tag{8}$$

where r_B is the radius of the Larmor orbit of an electron moving in a magnetic field of magnitude H. As a consequence, the field parameter, α, is just proportional to the strength of the magnetic field.

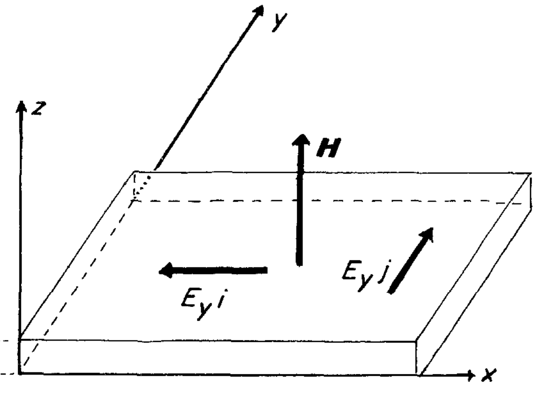

Figure 1 The geometry of the model.

Because the current is for the geometry of the model confined to the x axis, the Hall coefficient, R_{Hf}, and the electrical conductivity, σ_f, of the metal film are, respectively, defined by

$$R_{Hf} = \left.\frac{E_y}{HJ_x}\right|_{J_y=0} \tag{9}$$

$$\sigma_f = \left.\frac{J_x}{E_x}\right|_{J_y=0} \tag{10}$$

which then take the following forms

$$R_{Hf}/R_{H0} = \frac{2}{3}\,\frac{\mathscr{B}}{\mathscr{A}^2 + \alpha^2\mathscr{B}^2} \tag{11}$$

$$\sigma_f/\sigma_0 = \frac{3}{2}\left[\frac{\mathscr{A}}{\mathscr{A}^2 + \alpha^2\mathscr{B}^2}\right]^{-1} \tag{12}$$

where R_{H0} and σ_0 are, respectively, the Hall coefficient and the electrical conductivity of the bulk metal.

Finally the transverse magnetoresistance of a thin metal film can be calculated from the preceding equation by means of the formula

$$\frac{\Delta\varrho_f}{\varrho_f} = \frac{\varrho_f(H) - \varrho_f(0)}{\varrho_f(0)} \tag{13}$$

2.2. Presentation of theoretical results

Let us recall that neglecting formally the influence of the r.m.s. surface roughness the size effects in σ_f and R_{Hf} can be treated successfully by a Cottey method [18]. Equations 11, 12 and 13 remain valid in a Cottey analysis which yields analytical expressions for the functions $\mathscr{A}$ and $\mathscr{B}$

$$\begin{aligned}\mathscr{A}|_{\text{Cottey}} &= \mathscr{A}_c \\ &= \Big\{-\tfrac{1}{2}\mu + \mu^2 + \frac{\mu}{2}(1 - \mu^2 + \alpha^2\mu^2) \\ &\quad \times \ln\left[\frac{\alpha^2 + (1 + \mu^{-1})^2}{1 + \alpha^2}\right] \\ &\quad - 2\alpha\mu^3 \tan^{-1}\left[\frac{\alpha}{\mu}\left(\frac{1}{\alpha^2 + 1 + \mu^{-1}}\right)\right]\Big\}\end{aligned} \tag{14}$$

$$\begin{aligned}\mathscr{B}|_{\text{Cottey}} &= \mathscr{B}_c \\ &= \Big\{-\mu^2 + \mu^3 \ln\left[\frac{\alpha^2 + (1 + \mu^{-1})^2}{1 + \alpha^2}\right] \\ &\quad + \frac{\mu}{\alpha}(1 - \mu^2 + \alpha^2\mu^2) \\ &\quad \times \tan^{-1}\left[\frac{\alpha}{\mu}\left(\frac{1}{\alpha^2 + 1 + \mu^{-1}}\right)\right]\Big\}\end{aligned} \tag{15}$$

where the size parameter, μ, contains the constant specularity parameter, p

$$\mu = k\left[\ln\frac{1}{p}\right]^{-1} \tag{16}$$

Thus, as the field, thickness and roughness dependences of the film conductivity, magnetoresistance and Hall coefficient can be computed from Equations 12, 13 and 11, respectively, it is interesting to

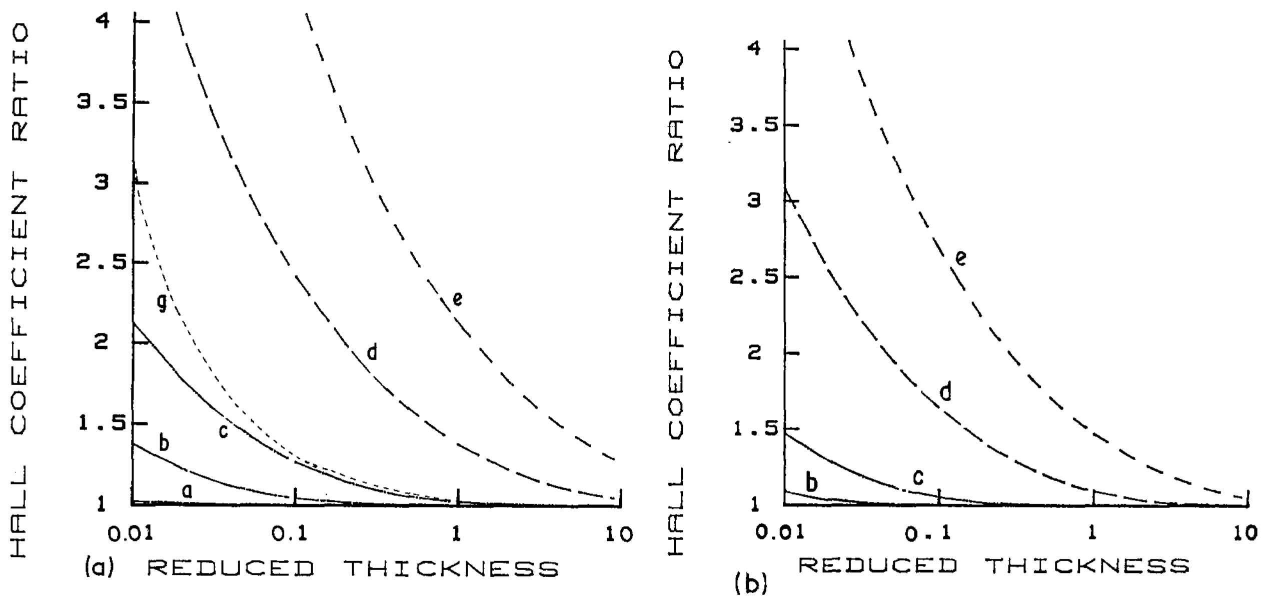

Figure 2 (a) Theoretical $R_{\mathrm{Hf}}/R_{\mathrm{H0}}$ against k plots for a moderate strength of the magnetic field ($\alpha = 1$); curves a, b, c, d, e are, respectively, for $r/\lambda_c = 0.01, 0.04, 0.1, 0.4$ and 1; curve g is the Cottey curve for $p = 0.5$. (b) The corresponding $R_{\mathrm{Hf}}/R_{\mathrm{H0}}$ against k plots in the limit of a strong magnetic field ($\alpha = 10$).

investigate how the angular dependence and the r.m.s. surface roughness modify the size dependence of these physical parameters.

First we have to consider the thickness dependence of these parameters. Fig. 2 shows the theoretical variation of the Hall coefficient ratio, $R_{\mathrm{Hf}}/R_{\mathrm{H0}}$, with the reduced thickness, k, for different values of the reduced r.m.s. roughness, r/λ_c, and for two fixed values of the magnetic field parameter, α. It is seen that the Hall coefficient decreases rapidly to 1 with increasing values of the reduced thickness whatever the strength of the magnetic field; such a result has been previously observed by some authors in the case of weak [17, 18] and strong [18] magnetic fields. The effect of the r.m.s. surface roughness seems to enhance the size effect in the reduced Hall coefficient. This behaviour is clearly depicted in Fig. 3; we note that as the external surfaces become more and more smooth (i.e. as r/λ_c tends to zero) the Hall coefficient of thin metal film tends to the bulk metal value.

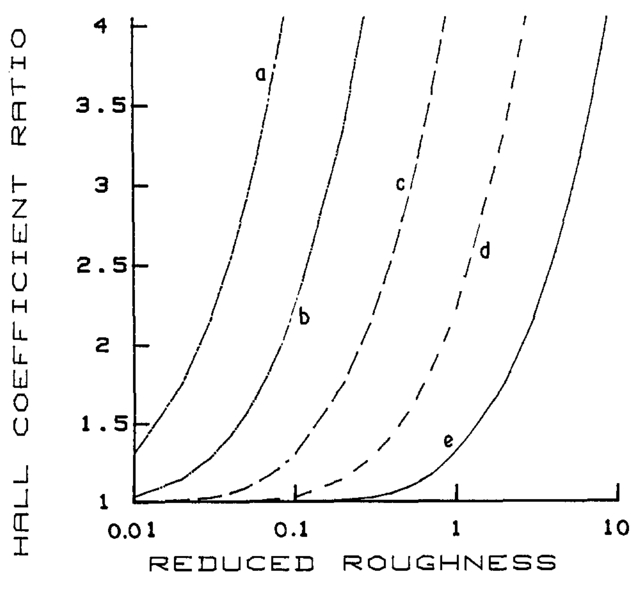

Figure 3 The surface roughness variation of the normalized Hall coefficient in the limit of small magnetic field ($\alpha = 0.1$) for different values of the reduced thickness, curves a, b, c, d and e are for $k = 0.001, 0.001, 0.1, 1$ and 10, respectively.

A comparison of the SC equations with the Cottey equations [18] is also possible. Effectively, if we assume that the main contribution to the current is due to electrons with θ in the range around $\pi/2$, the average value of $\cos^2\theta$ is taken to be $4/\pi^2$. Then returning to Equation 1 and substituting a constant value of 0.5 for the specularity parameter, p, we readily obtain a reduced r.m.s. roughness of about 0.104. Now comparing the Cottey curve (with a constant p equal to 0.5) with the SC curve for the r/λ_c value of 0.1 (Fig. 2 it clearly appears that the effect of the angular dependence is to decrease the apparent overall size effect in the Hall coefficient. A similar behaviour was previously reported by Tellier for the film conductivity in the absence of magnetic field [13].

Moreover from Table I it is seen that small oscillations in the Hall coefficient are nerver observed, even for very rough surfaces and strong magnetic fields. It

TABLE I Variation in the Hall coefficient with the product reduced thickness × field parameter: the case of a very thin film with $k = 0.01$

$k\alpha$	$r/\lambda_c = 0.1$, $R_{\mathrm{Hf}}/R_{\mathrm{H0}}$	$r/\lambda_c = 0.4$, $R_{\mathrm{Hf}}/R_{\mathrm{H0}}$
1	1.052090	1.672024
2	1.017204	1.428174
3	1.008249	1.316260
4	1.004782	1.249264
5	1.003105	1.203960
6	1.002174	1.171079
7	1.001605	1.146081
8	1.001233	1.097662
9	1.000976	1.110633
10	1.000792	1.097662
11	1.000655	1.086858
12	1.000551	1.077745
13	1.000470	1.069979
14	1.000405	1.063023
15	1.000353	1.057516

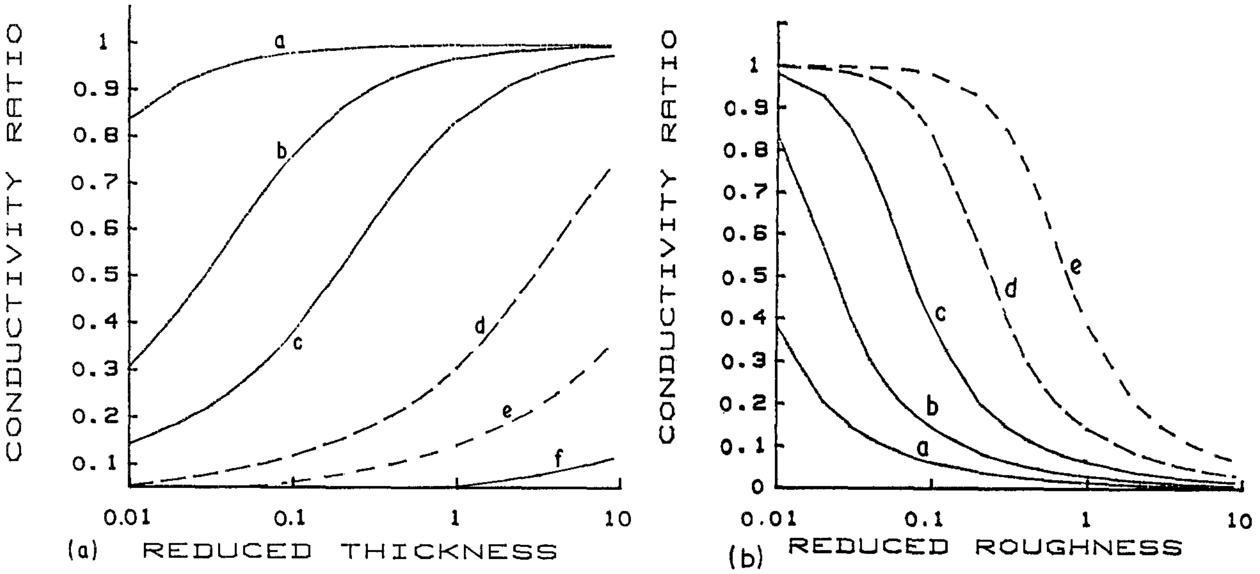

Figure 4 (a) Plots of the ratio σ_f/σ_0 against k for different values of r/λ_c and for $\alpha = 10$; curves a, b, c, d, e, f are, respectively, for $r/\lambda_c = 0.01$, 0.04, 0.1, 0.4, 1 and 4. (b) The σ_f/σ_0 against r/λ_c plots for $\alpha = 10$; curves a, b, c, d, e are for $k = 0.001$, 0.01, 0.1, 1 and 10, respectively.

should be noticed that in the framework of the Cottey model [18] very small oscillations are present in the Hall coefficient but the accuracy in the numerical evaluation which was accomplished with the aid of no more than a pocket calculator seems less firm when strong magnetic fields are applied in the z direction.

Plots of the ratio σ_f/σ_0 against k for different values of the r.m.s. reduced roughness and plots of σ_f/σ_0 against r/λ_c illustrated, respectively, in Fig. 4a and b exhibit the usual features: (1) the size effect vanishes for large reduced thicknesses; (2) transforming a smooth surface to a rough surface causes a marked decrease in the film conductivity ratio.

Thus, here we concentrate our attention to the theoretical variations of the reduced conductivity of the reduced roughness (Fig. 5). Let us recall that in the limit of small reduced thicknesses and in the case of diffuse scattering at the external surfaces Sondheimer [17, 19] and later Li and Marsocci [20] predicted oscillations of the conductivity which die out as the reduced thickness, k, and the constant specularity parameter, p, increase. In Fig. 5, the conductivity clearly presents no oscillations even for very thin films with rough surfaces ($r/\lambda_c = 0.2$) placed in strong magnetic fields (typically $\alpha \geqslant 10$). Thus here we disagree with previous theoretical works, but this disagreement concerns only the oscillating behaviour of the film conductivity with the strength of the applied magnetic field. Effectively, if we look to the variations of the conductivity ratio as a function of the field parameter, a crude analogy exists between the different theories which all satisfy the same essential physical requirements (see features 1 and 2 above). The discrepancies are only quantitative because we have previously demonstrated the size effect is less accentuated in the SC model [13, 16] than in Sondheimer [17] and Cottey [18] models.

Taking this last remark into account, one can reasonably expect smaller size effects in the transverse magnetoresistance when we treat the problem of the thin film resistivity in terms of the SC model than when we return to a Cottey formulation. Numerical

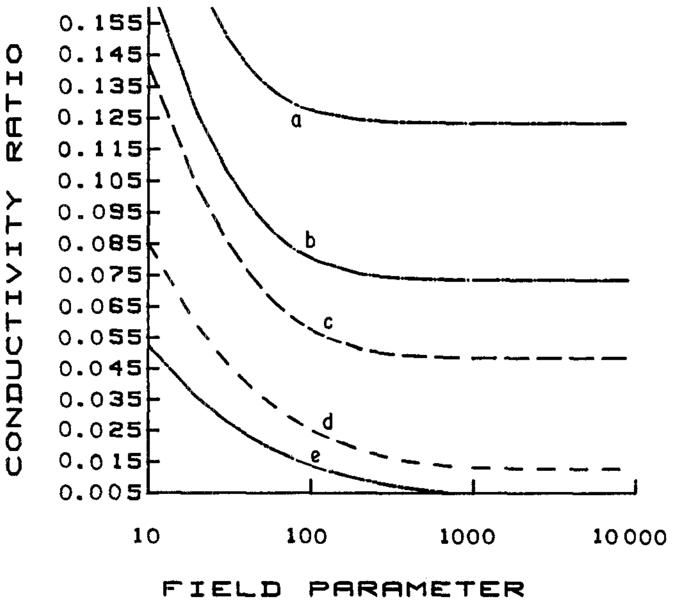

Figure 5 The field parameter variation of the conductivity ratio for a very thin film ($k = 0.01$) and for different values of the reduced roughness; curves a, b, c, d and e are for $r/\lambda_c = 0.06$, 0.08, 0.1, 0.2 and 0.4, respectively.

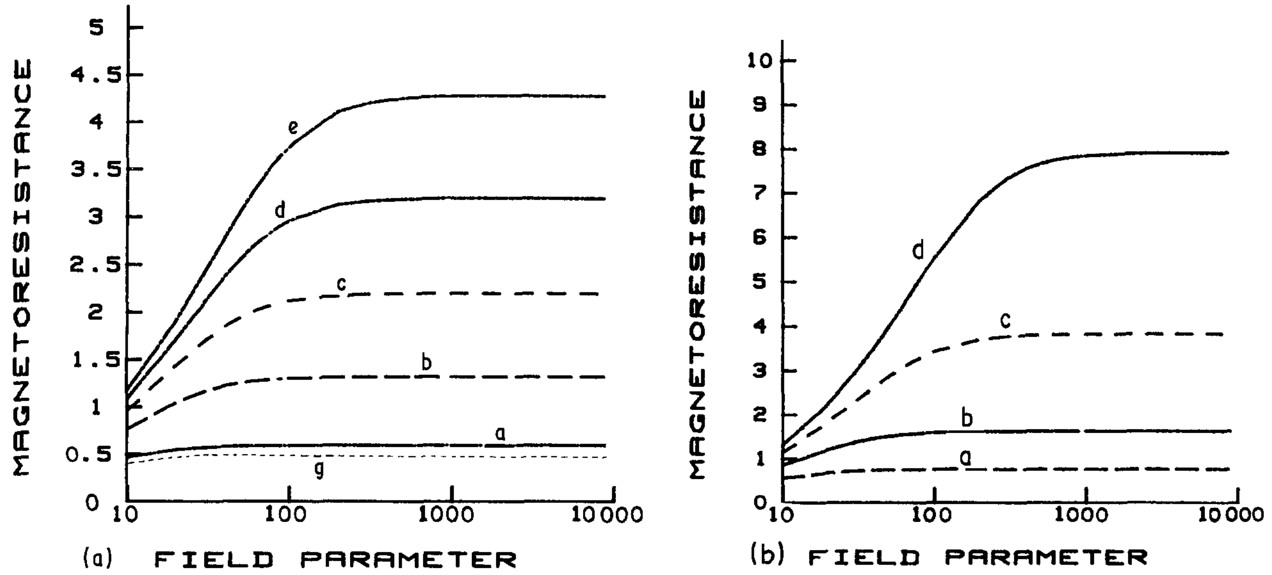

Figure 6 (a) The field parameter variation of the magnetoresistance for a very thin film ($k = 0.02$) and for different values of the reduced roughness; curves a, b, c, d and e are for $r/\lambda_c = 0.04, 0.06, 0.08, 0.1$ and 0.12, respectively; curve g is the Cottey curve for $p = 0.75$. (b) The results for $k = 0.1$; curves a, b, c, d are respectively for $r/\lambda_c = 0.1, 0.15, 0.25$ and 0.4.

computations (Table II) show that in reality the transverse magnetoresistance does not behave like other transport parameters (i.e. σ_f and R_{Hf}). The increase of this apparent magnetoresistance due, exclusively, to the limitation of the carrier mean free path by rough external surfaces, is particularly operative in the strong fields limit where the changes in the transverse magnetoresistance with the reduced thickness are found to be larger (about twice) with the SC model than with a Cottey formulation. The influence of the field parameter α on the transverse magnetoresistance is shown in Fig. 6 for different values of the r.m.s. surface. Curve g corresponds to the results of the Cottey formulation [18] for $p = 0.75$. A rapid comparison of curves b and g is sufficient to conclude that incorporating the r.m.s. surface roughness and the angular dependences in the calculation of the transverse magnetoresistance affects the field dependence in the same manner. As expected, the greater the α parameter, the more marked is the deviation between the predictions of the C model and the SC model.

Let us now undertake a systematic examination of the numerous theoretical results on the Hall coefficient and conductivity as computed from exact Equations 6 and 7 and from the following approximate equations

$$\mathscr{A} \simeq \frac{2}{3} - \frac{A}{12} + \frac{2A^2}{63} - \alpha^2\left(\frac{2}{3} - \frac{A}{4} + \frac{4A^2}{21}\right) \quad \alpha \ll 1 \qquad (17)$$

$$\mathscr{B} \simeq \frac{2}{3} - \frac{A}{6} + \frac{2A^2}{21} - \alpha^2\left(\frac{2}{3} - \frac{A}{3} + \frac{20A^2}{63}\right) \quad \alpha \ll 1 \qquad (18)$$

and

$$\mathscr{A} \simeq \frac{2}{3\alpha^2}\left(1 + \frac{A}{8}\right) \quad A \ll 1, \alpha \gg 1 \qquad (19)$$

$$\mathscr{B} \simeq \frac{2}{3\alpha^2} \quad A \ll 1, \alpha \gg 1 \qquad (20)$$

which hold, respectively, for small and large magnetic fields in order to define with certainty the validity of the approximate equations.

Tabulated values of the conductivity ratio, σ_f/σ_0, and of the normalized Hall coefficient, R_{Hf}/R_{H0}, for the case of weak magnetic fields (Table III) allows us to conclude that using Equations 17 and 18 instead of Equations 6 and 7 leads to satisfactory results (deviation less than 3%) in a larger k range ($k \geqslant 0.2$) for the conductivity than for the Hall coefficient. But numerical computations (Table IV) in the special case of strong magnetic fields, show that the validity of the approximate Equations 19 and 20 extends to a very large k range for both the conductivity and the Hall coefficient. Typically, for $\alpha = 40$ and for moderately rough external surfaces the deviation is less than 1% in the $k \geqslant 0.01$ range. Thus in the limit of strong magnetic fields the size effects in the electrical conductivity and the Hall coefficient can be conveniently described by means of the simple expressions [16]

$$\sigma_f/\sigma_0 \simeq \left(1 + \frac{A}{8}\right)^{-1} \quad A < 1, \alpha \gg 1 \qquad (21)$$

$$R_{Hf}/R_{H0} \simeq 1 \quad \alpha \gg 1 \qquad (22)$$

TABLE II Comparison of theoretical results as given in the framework of the Cottey model (C model) and the combined Soffer–Cottey model (SC model). The transverse magnetoresistance is evaluated from respective Equations 14 and 15 for $p = 0.5$ and from respective Equations 6 and 7 for $r/\lambda_c = 0.1$. Omitted values correspond to inaccuracies in the numerical evaluation

k	$\alpha = 0.04$		$\alpha = 4$		$\alpha = 40$	
	C model	SC model	C model	SC model	C model	SC model
0.07	9.79×10^{-5}	1.17×10^{-4}	2.15×10^{-1}	3.45×10^{-1}	4.00×10^{-1}	1.00
0.7	2.67×10^{-5}	3.90×10^{-5}	2.77×10^{-2}	5.75×10^{-2}	3.14×10^{-2}	7.08×10^{-2}
7	9.62×10^{-7}	1.90×10^{-6}	4.91×10^{-4}	1.33×10^{-3}	–	1.44×10^{-3}

TABLE III Comparison between the general expressions for the normalized Hall coefficient and for the reduced conductivity and the approximate expressions in the limit of small magnetic fields ($\alpha = 0.04$), assuming moderately rough surfaces ($r/\lambda_c = 0.04$)

k	Approximate equations		Exact equations	
	R_{Hf}/R_{H0}	σ_f/σ_0	R_{Hf}/R_{H0}	σ_f/σ_0
0.1	1.31108	0.90878	1.05488	0.82335
0.2	1.08206	0.91796	1.02241	0.88911
0.4	1.01740	0.94001	1.00789	0.93546
0.8	1.00359	0.96526	1.00245	0.96461
1	1.00236	0.97145	1.00164	0.97110
2	1.00055	0.98497	1.00046	0.98492
4	1.00013	0.99229	1.00012	0.99229
8	1.00004	0.99610	1.00003	0.99610

TABLE IV Comparison between the general expressions and the approximate expressions in the limit of large magnetic fields ($\alpha = 40$), assuming moderately rough surfaces ($r/\lambda_c = 0.04$)

k	Approximate equations		Exact equations	
	R_{Hf}/R_{H0}	σ_f/σ_0	R_{Hf}/R_{H0}	σ_f/σ_0
0.01	1.01432	0.25051	1.01142	0.24895
0.02	1 00333	0.39108	1.00308	0.39094
0.04	1.00081	0.55978	1.00079	0.55977
0.08	1.00020	0.71721	1.00020	0.71721
0.1	1.00013	0.76014	1.00013	0.76014
0.2	1.00003	0.86366	1.00003	0.86366
0.4	1.00001	0.92683	1.00001	0.92683
0.8	1.00000	0.96202	1.00000	0.96202
1	1.00000	0.96939	1.00000	0.96939
2	1.00000	0.98446	1.00000	0.98445
4	1.00000	0.99216	1.00000	0.99216
8	1.00000	0.99607	1.00000	0.99607

with a very sufficient accuracy until the term $(A + 1)/\alpha$ remains lower than 0.8.

Finally in this section we consider the dimensional effects in several transport parameters. Effectively it is now well known [1, 21–25] that to obtain a coherent set of physical parameters (e.g. the r.m.s. roughness, r/λ_c, the bulk parameter, σ_0, etc.) from experimental data, an extensive experimental investigation of several electrical and thermal properties in thin metal films is needed. Opting for this method to obtain an overall picture makes the understanding of the origin of the size effects less difficult, even if a systematic control of the morphology of films cannot be avoided. In the past the possibility of correlated size effects in various transport parameters have been studied [1, 26, 27] but theoretical information on this possibility is available only for models involving a constant specularity parameter. In particular, Tellier *et al.* considered the correlated effects in the Hall coefficient and in the product resistivity × temperature coefficient of resistivity (t.c.r.). Starting from the Cottey or the FS model they showed [26] that in the case of small magnetic fields the following general expression

$$R_{Hf}/R_{H0}|_{FS,C} \simeq \varrho_f\beta_f/\varrho_0\beta_0|_{FS,C} \quad \alpha < 1 \tag{23}$$

where ϱ_0 and β_0 refer, respectively, to the background resistivity and t.c.r. is valid.

In the absence of any magnetic field, evaluation of the electrical resistivity, ϱ_f, and of the temperature coefficient of resistivity, β_f, in terms of the SC model has revealed that the incorporation of both the surface roughness and the angular dependence in these parameters results in a decrease of the overall size effect which is considerably more marked for the film t.c.r. [28] than for the film conductivity. This behaviour can be regarded as singular with respect to behaviour predicted by other theories [1] which outline that the following relation

$$\beta_f\varrho_f \simeq \beta_0\varrho_0 \tag{24}$$

is satisfied in a very large k range. In this condition it becomes interesting to see if there are no consequences on the relationships between the Hall coefficient and the product resistivity × t.c.r. Comparing the tabulated values of the normalized Hall coefficient with those related to the product resistivity × t.c.r. (Table V) again opens up the possibility of correlating the size effects caused by external-surface scattering, in the Hall coefficient and in the product (resistivity, t.c.r.) because

$$R_{Hf}/R_{H0}|_{SC} \simeq \beta_f\varrho_f/\beta_0\varrho_0|_{SC} \quad \alpha < 1 \tag{25}$$

even if we are concerned with small reduced thicknesses or very rough surfaces for which large changes in the Hall coefficient with respect to the bulk are predicted.

3. Discussion

Because a systematic examination of the numerous experimental results [1, 21–25, 29–31] would be beyond the scope of this section, we shall restrict ourselves to two particular works [21, 25] for which the amount of experimental material seems sufficient to permit a reasonable interpretation. But before discussing these works it seems of interest to return to the feature which distinguishes the Sondheimer theory from the present model, namely the appearance of "Sondheimer" oscillations in the Hall coefficient and in the electrical resistivity with variation of the field strength and the "SC" monotonic change of the transport parameters with field. A tentative explanation of the origin of the oscillations has been made by Hurd [32] but he arrived at the conclusion that only some particular types of Fermi surface can give rise to oscillations in the galvanomagnetic properties. Remembering the Sondheimer model is based on the assumption of quasi free electrons we are no longer convinced by this explanation which applies solely to real metal films. Moreover, the matter published on this subject

TABLE V Comparison of the values of the product (reduced resistivity × reduced t.c.r.) calculated for $r/\lambda_c = 0.1$ with those of the normalized Hall coefficient calculated for $r/\lambda_c = 0.1$ in the case of a small magnetic field ($\alpha = 0.04$)

k	$\varrho_f\beta_f/\varrho_0\beta_0$	R_{Hf}/R_{H0}
0.001	4.42516	4.42466
0.004	2.89306	2.89275
0.01	2.22582	2.22560
0.04	1.57081	1.57068
0.1	1.30774	1.30766
0.4	1.09040	1.09036
1	1.03042	1.03040
4	1.00361	1.00361

is not wholly satisfactory and complete. Except for the work on aluminium films by Försvoll and Holwech [33], oscillatory behaviour is essentially observed in films of bismuth [1, 30, 34, 35], the only metal in which quantum size effects have been observed in the Hall effect [32]. If we turn to data on aluminium films we observe that for the thinner films the experimental amplitudes are smaller than predicted by theory; this discrepancy has been attributed to an imperfect treatment of the magnetoresistance. Effectively, Försvoll and Holwech used a modification of Kohler's rule to add the bulk magnetoresistance effect which does not vanish as predicted by the free electron model. But the oscillatory effect probably may be a "real metal" effect because aluminium presents a type of Fermi surface for which oscillations have been observed in the electrical resistivity [32].

In a study of the transport properties of well ordered bismuth films between 1.15 and 300 K, Hoffman and Frankl [25] reported measurements of the Hall effect, magnetoresistance and resistivity of bismuth films at different thicknesses in the range 0.07 to 3.7 μm. The crystallite size in these films is 5 to 10 μm and it is found to be practically independent of film thickness for thicknesses greater than 0.2 μm.

Bismuth is an highly anisotropic metal [36] and the galvanomagnetic transport parameters may be very sensitive to small differences in the number of electrons and holes as outlined by several authors [36, 37]. Thus the galvanomagnetic effects in bismuth crystals have been the object of calculations on the basis of the two-band model which leads to a non-vanishing magnetoresistance and to a temperature dependence of the Hall coefficient [37]. Let us note that Hoffman and Frankl observed, for T below about 20 K, a flattening in the temperature dependence of the Hall coefficient. Because one carrier conduction is characterized by a small coefficient which changes little with temperature [37], we can roughly assume that in the low-temperature region the bismuth film is exclusively p type and that size effects theories based on a single band conduction model can be used to interpret the thickness dependence of transport parameters. Several authors [25, 34, 38], among them Hoffman and Frankl, have also adopted this procedure to analyse the effect of the surface scattering on the resistivity, the Hall coefficient or other transport parameters of bismuth films, even if it is not wholly satisfactory. Thus we are now concerned with the size dependence of the resistivity and Hall coefficient of bismuth films at 4.2 K.

First, let us concentrate our attention on the thickness dependence of the resistivity. Hoffman and Frankl analysed the observed size effects in ϱ_f by using the following relation

$$\varrho_\mathrm{f} = \varrho_0\left[1 + \frac{3}{8k}(1 - p)\right] \qquad (26)$$

which constitutes down to $k \simeq 0.3$ an approximation of the general Fuchs–Sondheimer formula; accordingly a plot of the resistivity data in the form ϱ_f against k^{-1} yielded a value for ϱ_0 of about $0.31 \times 10^{-4}\,\Omega\,\mathrm{cm}$ and a value for the bulk mean free path (m.f.p.) λ_0 of about 8 μm. For film thicknesses, d, in the range 0.1 to 3.7 μm and for a bulk mean free path of about 8 μm it readily appears that Equation 26 is not still valid, so that the values for ϱ_0 and λ_0 determined by means of this equation are subject to question. With an average grain diameter in the range 5 to 10 μm it is reasonable to associate the scattering at grain boundaries with a grain parameter, ν, [1] in the range 1 to 2; thus the bulk grain-boundary resistivity will be about twice that of the perfect bulk metal. Surely, as suggested by Hoffman and Frankl, the scattering at the crystallite boundaries must give rise to a larger "bulk" resistivity than in a large single-crystal sample; but the difference between the observed value ($0.31 \times 10^{-4}\,\Omega\,\mathrm{cm}$) and the standard value ($3 \times 10^{-7}\,\Omega\,\mathrm{cm}$) is too large to be attributed solely to an additional scattering at the grain boundary. In reality this discrepancy can perhaps be due to an inadequate standard value. Effectively, Asahi and Kinbara [24] observed a small decrease in the background resistivity (about 10%) as the temperature changes from 77 to 4.2 K together with a value of ϱ_0 at 77 K close to that measured by Hoffman and Frankl. It should be pointed out that Hoffman and Frankl gave unsuitable estimates of ϱ_0 and λ_0 at 4.2 K, because they did not chose the approach of the Fuchs–Sondheimer theory [1]

$$\frac{\varrho_\mathrm{f}}{\varrho_0} = \frac{4}{3}\left(\frac{1 - p}{1 + p}\right)\frac{1}{k \ln 1/k} \qquad k \ll 1 \qquad (27)$$

commonly used for the limit of very small k; this implies the value of the background resistivity to be still larger. Consequently, taking into account the results reported earlier by Asahi and Kinbara, we assume that at 4.2 K the experimental value of the background resistivity is about $0.5 \times 10^{-4}\,\Omega\,\mathrm{cm}$; the appropriate value for the background mean free path is then determined by means of the formula

$$\varrho_0\lambda_0 \simeq \text{constant} \qquad (28)$$

We obtain $\lambda_0 \simeq 5\,\mu\mathrm{m}$. In this manner we are able to try to interpret the classical size effect in these bismuth films in terms of the SC model. Note that according to the SC model, the film conductivity, σ_f, for $k \ll 1$ is given by [15]

$$\ln\frac{\sigma_\mathrm{f}}{\sigma_0} \simeq \frac{1}{3}\ln k - \frac{2}{3}\ln\frac{r}{\lambda_\mathrm{c}} + \ln\frac{\pi}{\sqrt{3}} - \frac{2}{3}\ln 4\pi \qquad (29)$$

It must be pointed out that an $\ln \sigma_\mathrm{f}$ against $\ln d$ plot can yield neither the background resistivity nor the background mean free path; thus it remains necessary to assume values for ϱ_0 and λ_0. The corresponding $\ln \sigma_\mathrm{f}/\sigma_0$ against $\ln k$ plot is given in Fig. 7. A reasonable fit of the thickness dependence is observed for $k \lesssim 0.2$ yielding for the slope a value of about 0.33 which is close to the theoretical value of 1/3. A departure occurs for $k > 0.2$ showing that either the external surface becomes smoother as the film increases or the grain size varies with thickness. Values of the r.m.s. surface roughness as evaluated from Equation 29 are in the range 0.9 to 0.4.

If we wish to analyse the Hall coefficient data we are obliged, for lack of a standard value at 4.2 K, to determine an approximate value for $R_{\mathrm{H}0}$. Looking at

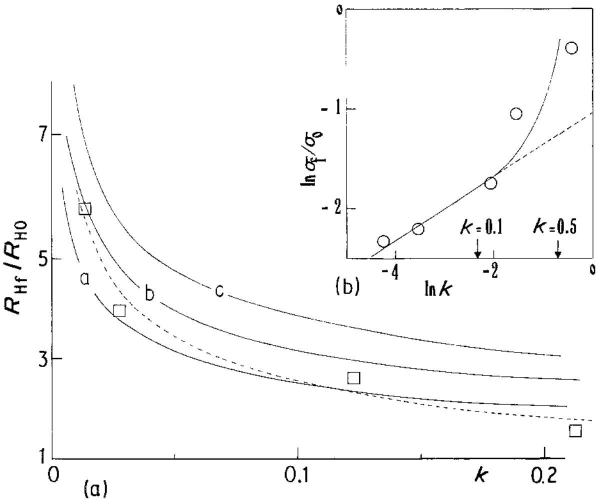

Figure 7 (a) Variation of the normalized Hall coefficient with the reduced thickness of bismuth films (Hoffman and Frankl [25]); full curves a, b, c at 4.2 K are the theoretical SC curves for $r/\lambda_c = 0.4$, 0.6 and 0.8, respectively. (b) The ln σ_f/σ_0 against ln k plot.

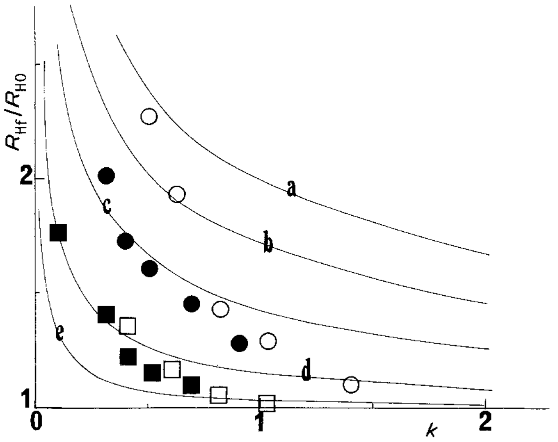

Figure 9 The reduced thickness dependence of the ratio R_{Hf}/R_{H0}; (○) measuring temperature (T_M) = 300 K and annealing temperature (T_A) = 373 K; (●) T_M = 80 K and T_A = 373 K; (□) T_M = 300 K and T_A = 523 K; (■) T_M = 80 K and T_A = 523 K. Curves a, b, c, d and e are the theoretical SC curves for $r/\lambda_c = 0.8$, 0.6, 0.4, 0.2 and 0.1, respectively.

the changes in R_{Hf} with temperature we see that the Hall coefficient is lowered by a factor of about 1.5 when the temperature increases from 4.2 to 80 K. Taking the standard value of $1.56 \times 10^{-9}\,\Omega\,\mathrm{cm}\,\mathrm{G}^{-1}$ for R_{H0} at 80 K the approximate value for R_{H0} at 4.2 K is found to be close to $2.4 \times 10^{-9}\,\Omega\,\mathrm{cm}\,\mathrm{G}^{-1}$. A tentative replotting of the Hall coefficient data in the form R_{Hf}/R_{H0} against k is illustrated in Fig. 7. We have also drawn in Fig. 7 theoretical SC plots corresponding to $r/\lambda_c = 0.4$, 0.6 and 0.8 to compare with the data. As we see, the best fit yields an r/λ_c value of about 0.5 in relative agreement with the value obtained from the thickness dependence of the resistivity. Although the choice of ϱ_0, λ_0 and R_{H0} values is arbitrary, we have roughly succeeded in interpreting the classical size effects in the transport parameters in terms of the SC model; surely another choice for the values of the bulk parameters can certainly lead to more satisfactory results. At this stage it does not seem fruitful to speculate about such possibilities. In practice it is proved that serious difficulties arise in the experimental determination of the bulk parameters when concerned with very small reduced thicknesses because the experimental plots related to resistivity data cannot yield separately the values of λ_0 and ϱ_0.

Let us now examine the case of transport properties of thin copper films [21]. Anomalously large size effects in R_{Hf} with respect to the predictions of the Fuchs–Sondheimer theory, were observed by Suri and coworkers in copper films. In particular, the thickness dependence at 300 and 80 K of the electrical resistivity and its temperature coefficient, the Hall coefficient, of thin copper films annealed at various temperatures have been extensively studied. These authors outlined the role played by a suitable dependence of frozen-in structural defects on the film thickness in determining the size effects. But turning to the changes in the ratio $\varrho_f\beta_f/\varrho_0\beta_0$ and in the normalized Hall coefficient with the film thickness we observe Fig. 8 a satisfactory coincidence between the $\varrho_f\beta_f/\varrho_0\beta_0$ and R_{Hf}/R_{H0} against d plots in agreement with the general formula [23]. As equation 23 is satisfied if the size effects are due to the electron scattering at film surfaces and/or at crystallite boundaries it seems of interest to reconsider the data for the Hall coefficient of copper films in the light of the SC model.

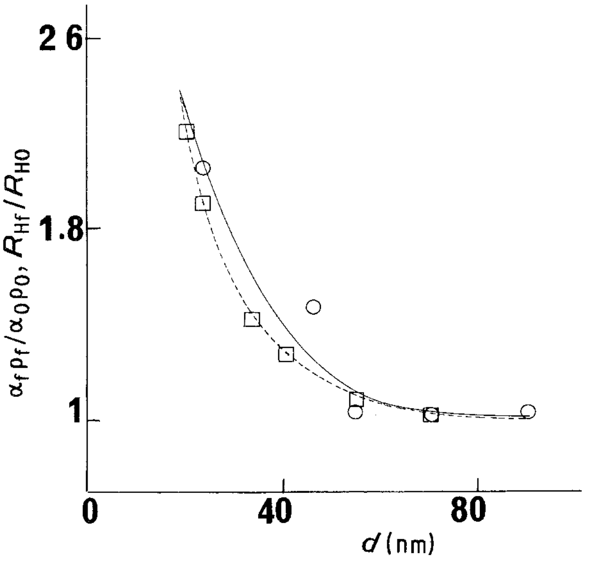

Figure 8 Thickness dependence of the ratios $\varrho_f\beta_f/\varrho_0\beta_0$ (○) and R_{Hf}/R_{H0} (□) at 300 K (Suri *et al.* [21]).

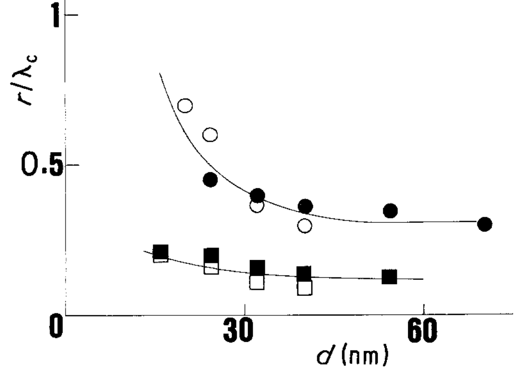

Figure 10 Variation of the r.m.s. surface roughness with thickness; (○) T_M = 300 K and T_A = 373 K; (●) T_M = 80 K and T_A = 373 K; (□) T_M = 300 K and T_A = 523 K; (■) T_M = 80 K and T_A = 523 K.

Certainly, among the frozen-in defects we have to consider the possible polycrystalline nature of copper films. However, Tellier [1] has previously shown that the Hall coefficient of an infinitely thick polycrystalline film is very close to that of a thick film free of grain boundaries. The Hall effect is thus not significantly affected by the grain-boundary scattering until this type of scattering is not the predominant scattering process; more precisely one can expect [39] a decrease in the Hall coefficient of less than 25% for the grain parameter, v, in the range $v \geqslant 1$.

Fig. 9 shows the reduced thickness dependence of $R_{\mathrm{Hf}}/R_{\mathrm{H0}}$ for copper films annealed at 373 and 523 K, assuming a bulk mean free path of about 38 nm [21] and 75 nm at, respectively, 300 and 80 K and a bulk Hall coefficient of about $5.5 \times 10^{-5}\,\mathrm{cm^3\,C^{-1}}$. The SC theoretical curves are also displayed in Fig. 9. By comparing with the theoretical curves we see that the observed variation of the normalized Hall coefficient can be understood only in terms of a thickness dependence of the surface roughness. The thickness variation of the r.m.s. surface roughness, as evaluated for copper films annealed at 373 and 523 K, is illustrated in Fig. 10. Data obtained from measurements of the Hall coefficient at 80 K are also used because for thicker films the inaccuracies of the determination of the normalized Hall coefficient and then of the r.m.s. surface roughness are less pronounced for measurements at 80 K than for measurements at 300 K. We observe that the r.m.s. surface roughness decreases rapidly with increasing values of film thickness to reach a limiting value which depends on the annealing temperature. This decrease is more marked at the lower annealing temperature than at the higher annealing temperature. From the limiting value of r/λ_c reported in Table VI we can obtain an average value of the specularity parameter by using the $|\cos\theta| \simeq 2/\pi$ relation. As a result we can conclude that successive annealings cause a reordering of the top surface of copper films; an interpretation usually advanced to explain the decrease of the film resistivity on annealing [1, 40–45].

At this point it must be remarked that analysing the resistivity data in terms of the SC model leads, in fact, to an enhancement of values of the r.m.s. surface roughness especially for thinner films (a deviation of about 30% is obtained for the thinnest film). This discrepancy may be attributed to the presence of crystallites whose average size varies with the film thickness. Because for $v \geqslant 1$, variations in the grain parameter, i.e. in the average grain diameter, do not induce marked changes in the Hall coefficient, the effects due to a thickness-dependent grain-boundary scattering process are masked when we study the thickness dependence of the Hall coefficient whereas they play an important role when we turn to the size dependence of the resistivity. To draw firm conclusions it is necessary firstly, to combine theoretically the SC model with the three-dimensional model of grain boundaries previously proposed by Tellier [46] and secondly to have quantitative informations on the changes in the mean crystallite size due to variations in film thickness and to subsequent annealing. Unfortunately, the experimental work of Suri *et al.* [21], though complete, does not give any information on the morphology of copper films. However, the grain-boundary scattering will roughly modify the size effect in R_{Hf}, ϱ_f and $\varrho_f\beta_f/\varrho_0\beta_0$ in the manner described above. Thus, our study strongly suggests that the observed behaviour of the size effect in transport properties on annealed copper films can be partly understood in terms of a surface re-ordering. The role played by grain boundaries in determining the size effect needs further theoretical study for obtaining valuable information. The SC model which leads to reasonable values for the r.m.s. surface roughness thus seems very interesting and convenient when it becomes necessary to undertake an investigation of the surface re-ordering on annealing.

4. Conclusion

Theoretical results in the presence of a transverse magnetic field are extensively presented for the electrical conductivity, the magnetoresistance and the Hall coefficient combining the Soffer and the Cottey models. Even in the regime of small thicknesses and strong magnetic fields the theoretical size effects in all transport parameters (e.g. ϱ_f, R_{Hf}, $\Delta\varrho_f/\varrho_f$) vary monotonically with the field parameter, α. Thus the oscillatory behaviour predicted by the Fuchs–Sondheimer theory is never observed. Incorporating surface roughness and angular dependence in calculations markedly diminishes the overall size effect in R_{Hf} and σ_f with respect to that predicted by theory, such as the Cottey model, involving a constant specularity parameter. Depature from this typical feature is observed for the magnetoresistance; the enhancement of the magnetoresistance with respect to the Cottey predictions is the result of the influence of the r.m.s. surface roughness. The theoretical variations in $R_{\mathrm{Hf}}/R_{\mathrm{H0}}$ and $\varrho_f\beta_f/\varrho_0\beta_0$ with the reduced thickness are found to exhibit a well-defined correlation with each other.

Attempts to fit previously published data, although nearly satisfactory, give evidence of some difficulty in interpretation arising as soon as we are concerned with the limit of very small reduced thicknesses (i.e. $k < 0.1$). This difficulty may be attributed to the possibility of determining separately ϱ_0 and λ_0 in the range $k < 0.1$; the value of r/λ_c obtained from data depends then on the choice of ϱ_0 and λ_0. In the regime $k > 0.1$, a combined Soffer–Cottey model seems as convenient as the Cottey or the Fuchs–Sondheimer

TABLE VI Variations in the limiting values of r/λ_c and in the corresponding average value of p with the annealing temperature, T_A

$T_A = 373\,\mathrm{K}$		$T_A = 523\,\mathrm{K}$	
Limiting value of r/λ_c	Average value of p	Limiting value of r/λ_c	Average value of p
0.31	0.002	0.12	0.40

models to analyse the size effects in galvanomagnetic properties of thin metal films provided the size dependences of various transport parameters (e.g. R_{Hf}, σ_f and β_f) measured simultaneously on the same metal films are available. In particular it becomes possible to follow roughly the changes in the r.m.s. surface roughness with the film thickness and with the annealing procedure by studying the variations of the Hall coefficient even if the electron scattering at grain boundaries cannot be completely neglected. However, care must be taken that a careful and consistent interpretation requires further systematic studies for obtaining quantitative information on the surface texture and on the film morphology.

References

1. C. R. TELLIER and A. J. TOSSER, "Size effects in Thin Films" (Elsevier, Amsterdam, 1982) Ch. 1 and 2.
2. K. L. CHOPRA, "Thin Film Phenomena" (McGraw-Hill, New York, 1969) Ch. 6.
3. T. J. COUTTS, *Thin Solid Films* **7** (1971) 77.
4. E. H. SONDHEIMER, *Adv. Phys.* **1** (1952) 1.
5. K. FUCHS, *Proc. Cambridge Phil. Soc.* **34** (1938) 100.
6. F. S. HAM and D. C. MATTIS, *I.B.M. J. Res. Dev.* **4** (1960) 143.
7. J. E. PARROTT, *Proc. Phys. Soc. London* **85C** (1965) 1143.
8. R. F. GREENE, *Phys. Rev.* **141** (1966) 687.
9. J. M. ZIMAN, "Electrons and Phonons" (Oxford University Press, London, 1962) Ch. IX.
10. S. SOFFER, *J. Appl. Phys.* **38** (1967) 1710.
11. J. R. SAMBLES and K. C. ELSOM, *J. Phys. D Appl. Phys.* **15** (1982) 1459.
12. J. R. SAMBLES, *Thin Solid Films* **106** (1983) 321.
13. C. R. TELLIER, *J. Mater Sci. Lett.* **3** (1984) 464.
14. A. A. COTTEY, *Thin Solid Films* **1** (1967–68) 297.
15. C. R. TELLIER, *J. Mater Sci.* **28** (1985) 4514.
16. *Idem, ibid.* **22** (1987) 2906.
17. E. H. SONDHEIMER, *Phys. Rev.* **80** (1950) 401.
18. C. R. TELLIER, M. RABEL and A. J. TOSSER, *J. Phys. F Metal Phys.* **8** (1978) 2357.
19. J. FEDER and T. JOSSANG, *Phys. Norv.* **1** (1963) 217.
20. T. C. LI and V. A. MARSOCCI, *Thin Solid Films* **12** (1972) 57.
21. R. SURI, A. P. THAKOOR and K. L. CHOPRA, *J. Appl. Phys.* **46** (1975) 2574.
22. S. KOCHOWSKI and A. OPILSKI, *Thin Solid Films* **48** (1978) 345.
23. H. SUGAWARA, T. NAGANO and A. KINBARA, *ibid.* **21** (1974) 33.
24. H. ASAHI and A. KINBARA, *ibid.* **66** (1980) 131.
25. R. A. HOFFMAN and D. R. FRANKL, *Phys. Rev. B* **3** (1971) 1825.
26. C. R. TELLIER, C. R. PICHARD and A. J. TOSSER, *J. Phys. F Metal Phys.* **10** (1980) L101.
27. C. R. TELLIER, L. HAFID and A. J. TOSSER, *J. Mater. Sci.* **15** (1980) 2991.
28. C. R. TELLIER, *ibid.* **22** (1987) 2043.
29. S. CHAUDHURI and A. K. PAL, *J. Appl. Phys.* **48** (1977) 3455.
30. V. P. DUGGAL and A. K. PAL, *ibid.* **40** (1969) 492.
31. V. P. DUGGAL and V. P. NAGPAL, *ibid.* **42** (1971) 4500.
32. C. M. HURD "The Hall Effect in Metals and Alloys" (Plenum, London, 1982).
33. K. FÖRSVOLL and I. HOLWECH, *Phil. Mag.* **9** (1964) 435.
34. M. INEOU, Y. TAMAKI and H. YAGI, *J. Appl. Phys.* **45** (1974) 1562.
35. N. GARCIA, Y. H. KAO and M. STRONGIN, *Phys. Rev. B* **5** (1972) 2029.
36. B. ABELES and S. MEIBOOM, *Phys. Rev.* **101** (1956) 544.
37. J. P. JAN, in "Solid State Physics", Vol. 5, Edited by F. Seitz and D. Turnbull (Academic, New York, 1959) pp. 1–98.
38. V. DAMORADAS and N. SOUNDARARAJAN, *Phys. Rev. B* **35** (1987) 5990.
39. C. R. PICHARD, C. R. TELLIER and A. J. TOSSER, *Thin Solid Films* **69** (1980) 157.
40. R. L. LONGBRAKE and S. J. BRIENT, *ibid.* **43** (1977) 343.
41. A. J. TOSSER, C. R. TELLIER and J. LAUNEY, *Vacuum* **27** (1977) 335.
42. C. R. TELLIER and A. J. TOSSER, *Thin Solid Films* **43** (1977) 261.
43. C. R. TELLIER, *Vacuum* **28** (1978) 321.
44. J. M. CHAUVINEAU and C. PARISET, *Surf. Sci.* **36** (1973) 1955.
45. R. E. HUMMEL and H. F. GEIER, *Thin Solid Films* **25** (1975) 335.
46. C. R. TELLIER, C. R. PICHARD and A. J. TOSSER, *ibid.* **61** (1979) 349.

Received 7 July
and accepted 9 October 1987