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Temperature coefficient of resistivity and deviation from Matthiessen's rule in a combined Soffer–Cottey model

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In the framework of a combined Soffer–Cottey model the temperature coefficient of resistivity is calculated by incorporating surface roughness and angular effects. In the limiting case of moderately rough surfaces a linear relation is proposed. Correlated size effects in the product of resistivity × temperature coefficient of resistivity are studied and vanishing of the deviation from Matthiessen's rule is predicted except for rough surfaces and for very thin films. Tentative attempts to fit previously published data on the basis of the present model are undertaken. Difficulties in controlling morphology and geometrical surface properties of films with various thicknesses are outlined. As a consequence a special emphasis is placed on procedures for overcoming these problems.

1. Introduction

Introduction of a constant specularity parameter, p, is a simple solution by means of which the development of theoretical models describing size effects in the conductivity of thin metal films, such as the well-known Fuchs–Sondheimer [1, 2] or Cottey [3] models, can be achieved. Among the many experiments on thin metal films graphical determinations of the specularity parameter were performed [4–6] from resistivity measurements. However previously published results on silver [7–9], copper [10, 11] and zinc films [12] do not agree very well with the predictions of these models even if the contribution of imperfections to transport properties seems to remain negligible [7, 8, 11, 12] at large thicknesses. Some authors [7, 8, 11, 12] advanced arguments to suggest that these discrepancies can be understood in terms of a thickness dependent specularity parameter.

Hence in the past few years interest in theories [13–18] relating the specularity parameter to the root mean square surface roughness, r, [13–17] or to the angle, θ, of incidence of the electron at the surface [15, 16, 18] has been revived. In particular some attempts have been made by Sambles and co-workers [19–22] to show that resistivity data can be better explained by the Soffer model [15] than the Fuchs–Sondheimer theory. Really the Soffer model presents the advantages to improve the Fuchs–Sondheimer model of surface scattering and to lead to a simple expression for the specularity parameter when the correlation length along the surface is taken to be zero. In effect a significant result is that in the case of uncorrelated surfaces the specularity parameter p which includes the influence of the r.m.s. surface roughness, r, and of the angle, θ, of incidence is simply expressed as [15]

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

where λ_c is the wavelength associated with the carrier. Recently combining with Cottey and Soffer models Tellier [23, 24] proposed an alternative method which offers analytical equations for the reduced film conductivity and allows an easy experimental determination [24] of the r.m.s. surface roughness in thin films.

However to discuss the validity of a model it is necessary to undertake a systematic comparison of the theoretical predictions of this model with the experimental results on various transport parameters. A critical analysis of the observed size effects is possible if at least the thickness dependence of the temperature coefficient of resistivity (t.c.r.) and of the conductivity were measured simultaneously. Thus the purpose of this paper is to derive a new analytical expression for the film t.c.r. in which the effects of the r.m.s. surface roughness and the general case of oblique incidence are included in order to reinterpret previously published data.

2. Analytical equation for the t.c.r.

Combining the Cottey and Soffer models the film conductivity, σ_f, is finally expressed in terms of the reduced film thickness, $k = d/\lambda_0$, and of the reduced roughness, r/λ_c. In effect, the general form for the reduced conductivity is

$$\sigma_f/\sigma_0 \;=\; F(\kappa) \qquad (2)$$

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with

$$F(\kappa) = \frac{\kappa}{2}\left[\frac{1}{2}\ln\left(\frac{(\kappa+1)^2}{\kappa^2-\kappa+1}\right) + 3^{1/2}\left(\tan^{-1}\frac{2-\kappa}{3^{1/2}\kappa} + \frac{\pi}{6}\right)\right] - \frac{1}{2A}\ln(1+A) \qquad (3)$$

where σ_0 is the background conductivity and the variables κ and A are given by

$$\kappa = A^{-1/3} \qquad (4)$$

$$A(r, k) = \frac{1}{k}\left(\frac{4\pi r}{\lambda_c}\right)^2 \qquad (5)$$

Considering the following usual assumptions [6, 25]

1. The rigid band model is valid
2. The number of conduction electrons per unit volume is temperature independent in the experimental range
3. The thermal expansion of the film thickness, d, is negligible with respect to that of the background mean free path, λ_0
4. The r.m.s. surface roughness remains unaffected by thermal variations as expected for well-annealed metal films

which are generally retained to interpret data on film t.c.r. and further neglecting the thermal expansion mismatch between the film and its substrate, the logarithmic differentiation of Equation 2 gives

$$\frac{d\sigma_f}{\sigma_f} = \frac{d\sigma_0}{\sigma_0} + \frac{dF(\kappa)}{F(\kappa)} \qquad (6)$$

Since the differential of $F(\kappa)$ can be expressed as

$$dF(\kappa) = \frac{d\kappa}{\kappa}[F(\kappa) + H(\kappa)] \qquad (7)$$

with

$$H(\kappa) = -\frac{3}{2A}\left[\frac{1}{(\kappa+1)(\kappa^2-\kappa+1)} - \frac{A}{1+A} + \frac{2}{3}\ln(1+A)\right] \qquad (8)$$

Equation 6 can be transformed into

$$\frac{d\sigma_f}{\sigma_f} = \frac{d\sigma_0}{\sigma_0} + \frac{1}{F(\kappa)}\frac{d\kappa}{\kappa}[F(\kappa) + H(\kappa)] \qquad (9)$$

Turning to Equations 4 and 5 it follows that

$$3\frac{d\kappa}{\kappa} = -\frac{d\lambda_0}{\lambda_0} \qquad (10)$$

Inserting Equation 10 into Equation 9 and taking into account that under assumptions 1 and 2 the bulk t.c.r. is

$$\beta_0 = -\frac{1}{\sigma_0}\frac{d\sigma_0}{dT} = -\frac{1}{\lambda_0}\frac{d\lambda_0}{dT} \qquad (11)$$

the final expression for the film t.c.r., β_f, which is defined as

$$\beta_f = -\frac{1}{\sigma_f}\frac{d\sigma_f}{dT} \qquad (12)$$

becomes

$$\beta_f = \beta_0\left[1 - \frac{1}{3}\left(1 + \frac{H(\kappa)}{F(\kappa)}\right)\right] \qquad (13)$$

At this point it should be noticed that t.c.r. measurements are generally performed at temperatures close to room temperature [7, 8, 10–12, 26–32] so that for continuous films of the most common metals the reduced thickness does not take values lower than 0.2. Thus if the reduced roughness does not exceed 0.04 the dimensional parameter κ remains larger than unity. In these conditions it may be of interest to derive an asymptotic expression for the film t.c.r. in the limiting case of high values of the parameter κ.

Expanding Equation 8 in the power series of reciprocal κ and further neglecting terms of power higher than four gives the $H(\kappa)$ function in the form

$$H(\kappa) \simeq \frac{1}{2\kappa^3} - 1, \qquad \kappa > 1 \qquad (14)$$

Since in the limit of large κ the conductivity ratio reduces to [24]

$$\frac{\sigma_f}{\sigma_0} \simeq 1 - \frac{1}{8\kappa^3}, \qquad \kappa > 1 \qquad (15)$$

the t.c.r. ratio is readily found to be

$$\frac{\beta_f}{\beta_0} \simeq 1 - \frac{1}{8\kappa^3}, \qquad \kappa > 1 \qquad (16)$$

that is just the result that one can easily obtain using the approximate Equation 15 for the film conductivity.

3. Theoretical results

3.1. The reduced t.c.r.

Equations 2 and 13 can be evaluated numerically with the aid of a pocket calculator, but for convenience the theoretical curves presented in this section are drawn using a microcomputer. Fig. 1 illustrates for different values of the reduced roughness the limitation of the bulk mean free path by the film surfaces. To make the comparison with the Cottey model more easy the variation of the t.c.r. ratio as a function of the reduced thickness are also shown in Fig. 2 for two values of the constant specularity parameter. In Fig. 3 the dimensionless reduced t.c.r., β_f/β_0, is plotted against the r.m.s. reduced roughness, r/λ_c, according to Equation 13, the reduced thickness, k, acting as a parameter.

Several points arising from these figures merit some comments

1. The t.c.r. ratio exhibits a thickness dependence in accord, at first sight, with other size effect theories. However turning our attention (Fig. 2) to the Cottey curves and to the curve obtained for $r/\lambda_c = 0.06$ (i.e. for $p(r, \theta) \simeq 0.57$ at normal incidence) it is clear that the incorporation of both the surface roughness and the angular dependence in this transport parameter results in a serious decrease of the overall size effect.

2. For the reduced thickness in the range 0.1 to 10 the reduced t.c.r. is dominated by the surface roughness. However as expected the size effect vanishes in

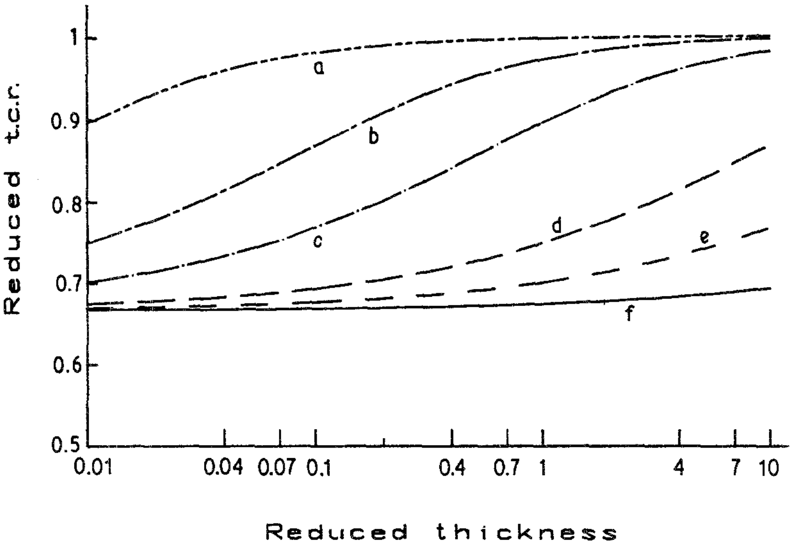

Figure 1 The reduced t.c.r., β_f/β_0, against the reduced thickness, k, for different values of the reduced roughness, r/λ_c. a, b, c, d, e, f: theoretical curves for the respective r/λ_c values of 0.01, 0.04, 0.1, 0.4, 1 and 4.

the limit of very small r/λ_c, i.e. when the film surface has zero roughness.

3. With increasing reduced roughness ($r/\lambda_c > 0.4$) the thickness dependence of the t.c.r. ratio has a tendency to be masked by the very predominant surface roughness effect and it seems that the film t.c.r. cannot reach the bulk t.c.r. even for very thick films. But in a preceding paper [23] an examination of numerical results for the reduced conductivity obtained by extending either the Cottey [23] or the Fuchs–Sondheimer [19] theory has revealed that the range of applicability of Equation 2 extends to about $r/\lambda_c \simeq 0.2$. Since to our knowledge the combined Fuchs–Sondheimer and Soffer model has not yet been utilized to derive theoretical expressions for transport parameters other than the reduced conductivity it is more realistic in the absence of contradictory information to admit that the same range of applicability holds also for Equation 13.

3.2. The product resistivity × t.c.r.

Because the product of the resistivity and the electronic mean free path should remain constant [33, 34] the validity of the Fuchs–Sondheimer theory was questioned by several authors [35–37]. In general this problem was treated in terms of the Matthiessen's rule which states that the resistivity, ϱ_f, can be calculated by superimposing the contributions to resistivity due to all the sources of electronic scatterings, i.e.

$$\varrho_f = \varrho_{ph} + \varrho_S + \varrho_I \qquad (17)$$

where ϱ_S represents the resistivity due to scattering at external surfaces, ϱ_I is the residual resistivity due to impurities or frozen-in defects and ϱ_{ph} is the ideal phonon resistivity. Among these resistivity terms only the contribution ϱ_I is not temperature dependent so that it is possible to rewrite the surface resistivity in the form [37]

$$\varrho_S(T) = \varrho_S^*(0) + \Delta\varrho_S(T) \qquad (18)$$

where $\varrho_S^*(0)$ is the residual resistivity due to surface scattering and $\Delta\varrho_S(T)$ can be regarded as the deviation from Matthiessen's rule on the surface scattering (referred as DMR hereafter).

An evident method to evaluate the DMR is to start from the theoretical formulae for the surface resistivity:

$$\varrho_S/\varrho_0 = \frac{1 - F(\kappa)}{F(\kappa)} \qquad (19)$$

derived from Equation 2 expressing the film conductivity on the basis of the Soffer–Cottey model.

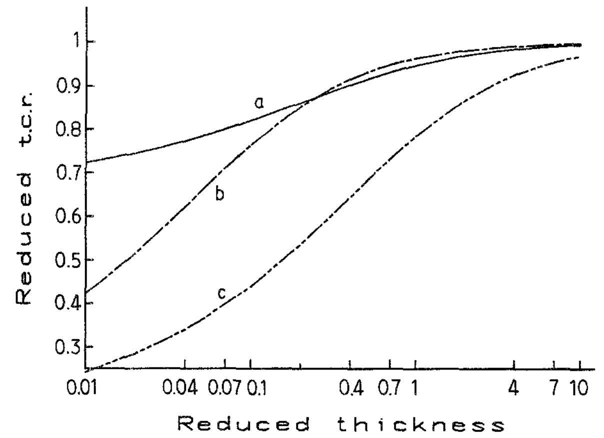

Figure 2 Comparison of the present model (curve a, $r/\lambda_c = 0.06$) with the Cottey model (curves b and c for the respective p values of 0.9 and 0.4).

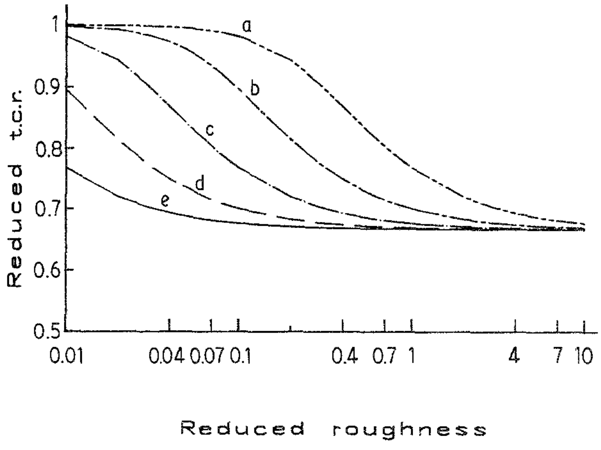

Figure 3 The reduced t.c.r., β_f/β_0, against the reduced roughness, r/λ_c, for different values of the reduced thickness, k. a, b, c, d, e: theoretical curves for the respective k values of 10, 1, 0.1, 0.01 and 0.001.

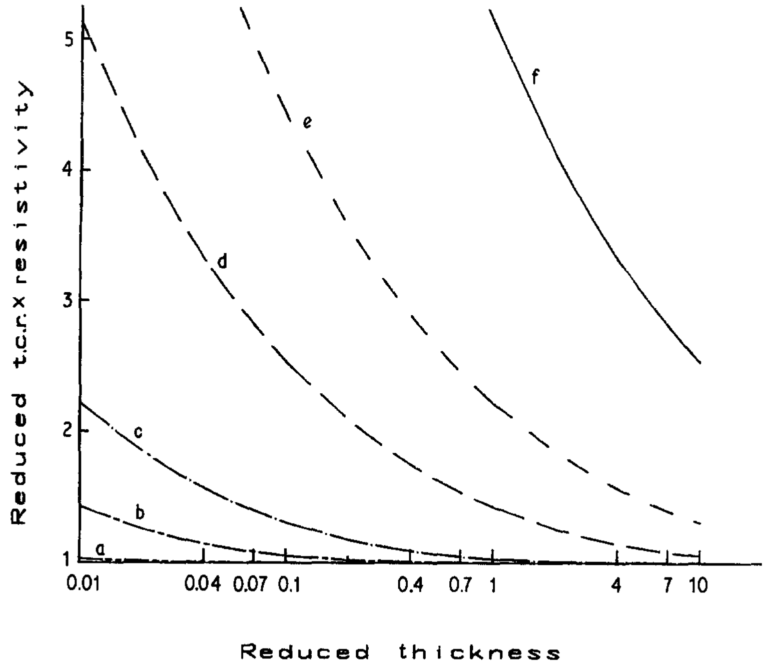

Figure 4 The reduced product, $\beta_f\varrho_f/\beta_0\varrho_0$, against the reduced thickness, k, for different values of the reduced roughness, r/λ_c. a, b, c, d, e, f: theoretical curves for the respective r/λ_c values of 0.01, 0.04, 0.1, 0.4, 1 and 4.

Alternatively, the total film resistivity ϱ_f is expressed as

$$\varrho_f(T) = \varrho_{ph}(T) + \varrho_i(0) + \varrho_S(T) \quad (20)$$

so that partially differentiating Equation 20 with respect to temperature, T, gives

$$\beta_f\varrho_f = \varrho_0\beta_0 + \varrho_S\beta_S \quad (21)$$

with, for bulk parameters

$$\varrho_0 = \varrho_{ph} + \varrho_i, \qquad \varrho_0\beta_0 = \varrho_{ph}\beta_{ph} \quad (22)$$

Combining Equations 18 and 21 we have

$$\beta_f\varrho_f = \varrho_0\beta_0 + \Delta\varrho_S\Delta\beta_S \quad (23)$$

Since in the framework of the combined Soffer–Cottey theory the asymptotic expression of the surface resistivity in the limit of large κ is [24] (see Equation 15)

$$\varrho_S(T) \sim \varrho_0(T)\lambda_0(T), \qquad \kappa > 1 \quad (24)$$

that is to say that the surface resistivity $\varrho_S(T)$ reduces to a temperature independent term, i.e. to the residual surface resistivity, it is clear that the only equation expressing the Matthiessen's rule is

$$\beta_f\varrho_f = \varrho_0\beta_0 = \varrho_{ph}\beta_{ph} \quad (25)$$

and that we identify the product $\Delta\varrho_S\Delta\beta_S$ with a DMR. Hence an alternative and convenient approach to the DMR for very thin films ($k < 1$) consists of studying the variations in the product resistivity × t.c.r.

The theoretical results are displayed in Figs 4 to 6. Fig. 4 clearly reveals that in the usual range of applicability of Equations 2 and 13 the reduced product resistivity × t.c.r. approaches unity as soon as the reduced thickness takes values greater than 0.4. The major effect when considering the surface roughness and the angular dependence of the specularity parameter seems to diminish the variation of the product resistivity × t.c.r. with the film thickness, effectively taking a constant specularity parameter causes an

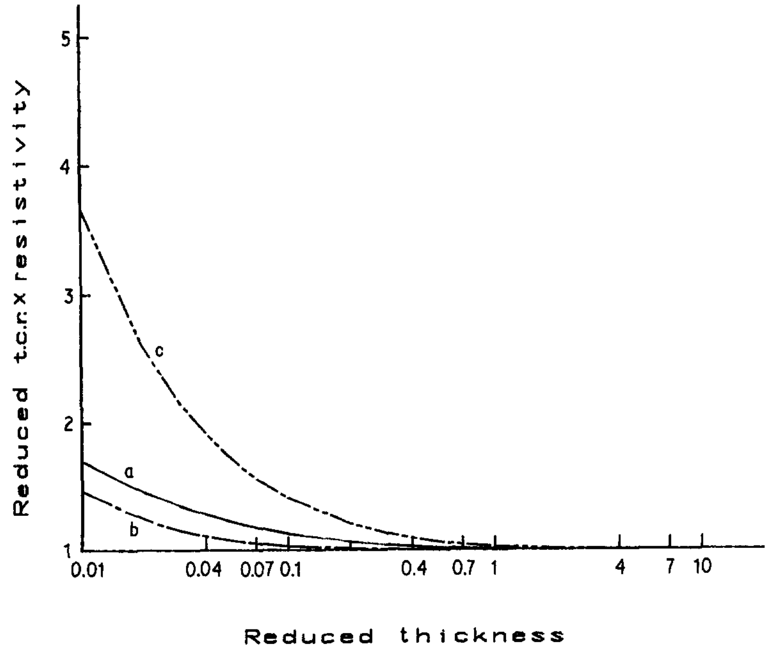

Figure 5 Comparison of the present model (curve a, $r/\lambda_c = 0.06$) with the Cottey model (curves b and c for the respective p values of 0.9 and 0.4).

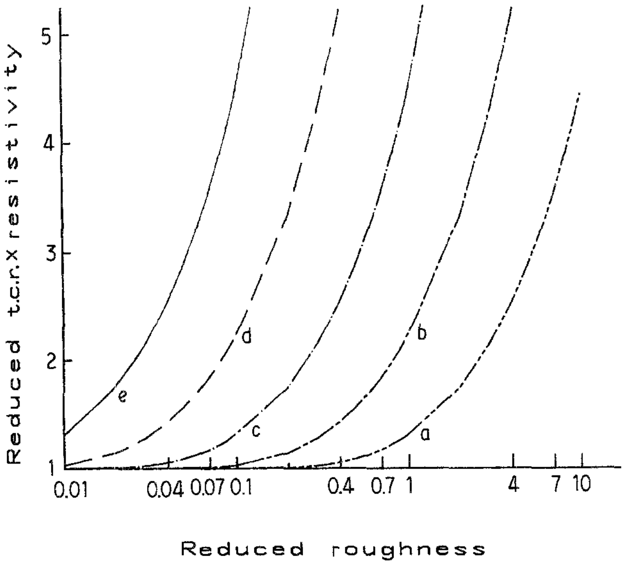

Figure 6 The reduced product, $\beta_f\varrho_f/\beta_0\varrho_0$, against the reduced roughness, r/λ_c, for different values of the reduced thickness, k. a, b, c, d, e: theoretical curves for the respective k values of 10, 1, 0.1, 0.01 and 0.001.

increasing size effect (Fig. 5). The apparent decrease in the DMR is also evidenced by the Fig. 6 which shows that for relatively thick films the reduced surface roughness must reach the large value of unity to observe a marked DMR.

4. Discussion

In the following we first discuss previous published data on film t.c.r. in terms of the present model. Because of difficulty in controlling the bulk properties of films in a large domain of thickness this section emphasises only a few experimental works in order to consider some problems associated with a theoretical interpretation of t.c.r. data. Then the theoretical results for the DMR are used to try to reinterpret the almost complete and comprehensive study of Nakamichi and Kino on aluminium strips [37].

Most of size effects in the t.c.r. of thin films have been analysed in terms of the Fuchs–Sondheimer theory by plotting the data in the form $1/\beta_f$ against $1/d$ or $\beta_f d$ against d [3–6, 10, 28, 30, 38]. The Fuchs–Sondheimer or the Cottey model effectively predicts that straight line relations can represent the thickness dependence accurately down to $k \simeq 0.3$. Such fits yield both the infinitely thick film t.c.r. β_0 and the term $(1 - f)\lambda_0$ (Fuchs–Sondheimer model) or $\lambda_0/\ln(1/p)$ (Cottey model). Obtaining a linear t.c.r. plot is in most cases interpreted as an adequacy of the Fuchs–Sondheimer theory to explain the observed size effect. To demonstrate how trivial this conclusion is let us examine the approximate Equation 16. It is clear that for relatively thin films ($k > 0.4$) with moderately rough surfaces ($r/\lambda_c < 0.06$) a straight line behaviour is also predicted by the present model. Hence finding a simple linear d (or $1/d$) dependence in $\beta_f d$ (or in $1/\beta_f$) is not necessarily a consequence of a constant specularity parameter. This point is very important since grain-boundary models can also lead to straight line relationships when modelling the simultaneous electronic scattering at the external surfaces and at the grain-boundaries [6]. Thus apparently, when the resistivity and the temperature coefficient of resistivity were measured in a large thickness range, the only approach to decide the validity of the present model is to compare the overall size effects in film resistivity and t.c.r. If this overall size effect is less accentuated for the t.c.r. data than for the resistivity data the applicability of theories involving a constant specularity parameter can be questioned.

Among the various works [7–12] in which an apparent dependence of the specularity was observed and thus which can be understood in terms of the combined Soffer–Cottey model several [7, 8, 11, 12] exhibit size effect in the t.c.r. which are too pronounced for very thin films ($d < 30.0$ nm) to be interpreted only in terms of a surface roughness effect. The lack of information on the film structure [7, 11] or on the values of the infinitely thick film resistivity and t.c.r. [11, 12] does not enable us to establish the real cause of such enhanced size effects: for example, a semi-continuous nature of films, the existence of a dependence on thickness of a large concentration of frozen in point defects, a dominant electronic scattering at grain boundaries. However some experiments [31, 38, 39] reveal differences in the overall size effect in film resistivity and film t.c.r. It is in particular the case for the work by Ghosh and Pal [38] on evaporated nickel films, unfortunately the dispersion of their t.c.r. data is not appropriate for a meaningful analysis.

Hence we have only selected two experimental works to undertake a comparison with the Soffer–Cottey model. First we are concerned with measurements of the resistivity and temperature coefficient of resistance made on annealed copper films by Leonard and Yu [31]. However these experimental results require some attention because values of the reduced t.c.r. of very thick films are too low with respect to those of the reduced conductivity causing the reduced product $\varrho_f\beta_f/\beta_0\varrho_0$ to take physically unreasonable values, i.e. values smaller than unity. Assuming that the measured values of film resistivity are correct since in general some difficulty can arise in the estimation of film t.c.r. (see for example assumptions 3 and 4 in Section 2) a correcting factor is applied to the bulk t.c.r. The corrected data together with data on gold films [39] are reported in Fig. 7. Examination of Fig. 7 reveals some slight deviation from the theory. A satisfactory fit of the data for gold films is obtained for $r/\lambda_c \simeq 0.08$. Since effect in t.c.r. of copper films is found to be connected with relatively high values of the reduced roughness in the range 0.1 to 0.16. These works illustrate the difficulties in interpreting t.c.r. data. These difficulties can have various origins, in particular they can be attributed to errors in the evaluation of films t.c.r. such as,

1. The misleading of the correction factor due to the effect of expansion mismatch on the temperature coefficient of resistance of thin films which can reach $4 \times 10^{-5}\,\mathrm{K}^{-1}$ for typical substrates [40].
2. The use of partially annealed films for which a continuous evolution of the film t.c.r. occurs with increasing temperatures.

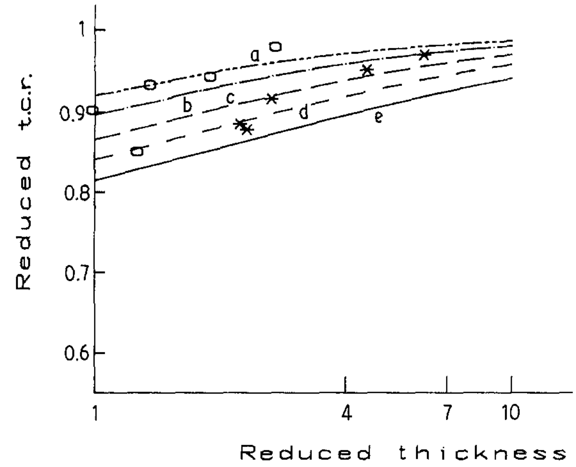

Figure 7 Theoretical fit of published data on (*) copper films [31] and (O) on gold films [39]. a, b, c, d, e theoretical Soffer–Cottey curves for the respective r/λ_c values of 0.08, 0.1, 0.13, 0.16 and 0.2.

3. The impossibility of knowing the true temperature of thin films, the temperature being in general measured by using thermal sensors placed on the substrate.

At large thickness small errors in the evaluation of temperature or correction factor can evidently cause some departures from the expected variation of the film t.c.r. with the film thickness.

But there is a more serious problem that is the independency of the geometrical properties of the upper film surface on the film thickness. Without doubt the morphology of films varies with thickness and there is no reason for the surface roughness to remain unaffected. From a crude point of view it seems that the surface roughness may be more accentuated in thin films than in thick films but only an extensive experimental study of the film flatness (scanning electron microscopy with backscattered electrons for example) would give some idea on this point.

The recent work by Nakimichi and Kino [37] on DMR is from this point of view very interesting because these authors studied aluminium strips over a large temperature range (1.5 to 60 K). Large variations in the reduced thickness ($k \simeq 0.02$ to 50) are thus obtained which permit one to investigate the size effect on a specimen and thus to overcome the difficulty of assigning precise surface roughness properties to various thin specimens. Without ambiguity the resistivity, $\varrho_S(T)$ due to surface scattering present a hump around 25 K when plotted against temperature. This behaviour is accentuated for the thinnest specimens. Nakimichi and Kino attempted to explain their results in terms of the Fuchs–Sondheimer model using the theoretical formulae [37]

$$\varrho_S = \varrho_f - \varrho_0 = \frac{\varrho_0 \lambda_0}{d} k[\mathscr{F}_{FS}(k, p) - 1] \quad (26)$$

where $\mathscr{F}_{FS}(k, p)$ is the function describing the size effect in resistivity on the basis of Fuchs–Sondheimer theory. For a given specimen $\varrho_0\lambda_0/d$ must remain constant with increasing temperature, thus variations in the term $k\ [\mathscr{F}_{FS}(k, p) - 1]$ with temperature are expected to show a qualitative agreement with experimental results.

Turning now to the combined Soffer–Cottey model the term $k\ [\mathscr{F}_{FS} - 1]$ must be replaced by

$$k\frac{\varrho_S}{\varrho_0} = k\left(\frac{\varrho_f}{\varrho_0} - 1\right) = k\left[\frac{1 - F(\kappa)}{F(\kappa)}\right] \quad (27)$$

and a theoretical representation of the DMR on the surface scattering is now obtained by plotting the reduced product surface resistivity × thickness against the reduced thickness (Fig. 8). We never see the maximum around $k = 0.8$ predicted by the Fuchs–Sondheimer theory even for relatively rough surfaces (i.e. $r/\lambda_c \simeq 0.15$ corresponding to $p \simeq 0.17$ for an oblique incidence of 45°). This is not surprising as the Cottey model for constant p in the range 0.4 to 0.9 does not evidence any hump in the $k\varrho_S/\varrho_0$ against k plots (Fig. 9) and that for $p = 0.5$ the hump observed in the Fuchs–Sondheimer model remains attenuated. This point is one of the reasons for which these authors have retained a constant value of zero for the specularity parameter, the second being that for $p = 0$ the product $\varrho_0\lambda_0$ takes a value ($\simeq 1.1 \times 10^{-3}\,p\Omega\text{m}^2$)

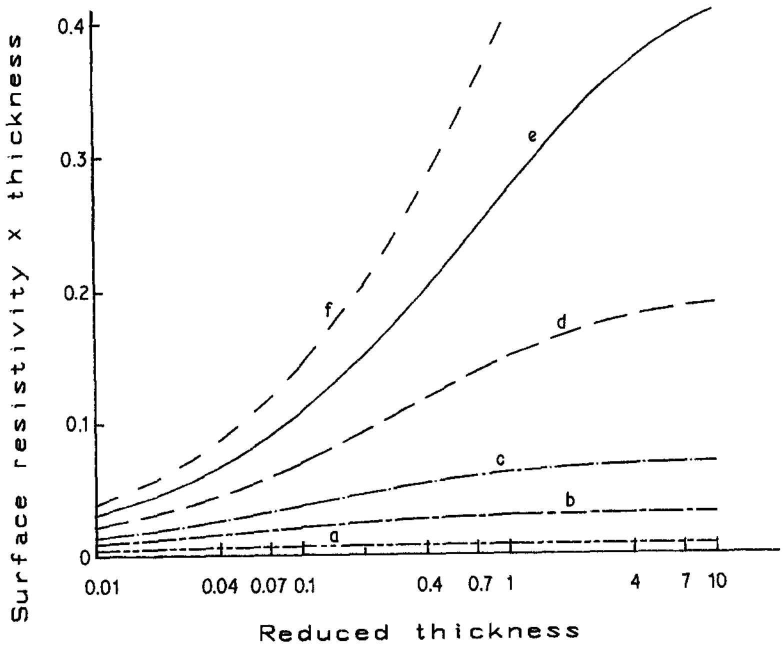

Figure 8 A plot of $k\varrho_S/\varrho_0$ against k for different values of r/λ_c. a, b, c, d, e, f: theoretical curves for the respective r/λ_c values of 0.02, 0.04, 0.06, 0.1, 0.15, 0.2.

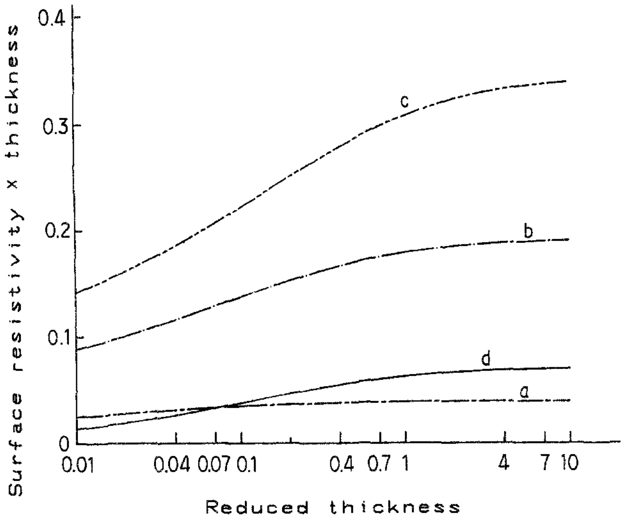

Figure 9 A Cottey plot of $k\varrho_S/\varrho_0$ against k. a, b, c: theoretical Cottey curves for the respective p values of 0.4, 0.6 and 0.9. D is the theoretical Soffer–Cottey curve for $r/\lambda_c = 0.06$.

which is the closest to that ($4 \times 10^{-4} p\,\Omega\mathrm{m}^2$) calculated by the free electron theory. Using a constant value of $\varrho_0\lambda_0$ in the theoretical Fuchs–Sondheimer calculation the consistency between the theoretical curves and the experimental ones is satisfactory only at low temperatures i.e. for small k. Serious departure are observed for all specimens as soon as k reaches a value of about 2 (Fig. 6, [37]). Note also that the Soffer–Cottey results depicted in Fig. 8 should certainly fit the data well since for the thinnest aluminium strip the experimental points are situated below the Fuchs–Sondheimer theoretical ones for $k < 0.1$ and that the inverse situation is observed for $k > 0.1$. Such a behaviour is consistent with the Cottey–Soffer model (compare for example Curves B and D in respective Figs 8 and 9).

To partially suppress these discrepancies a tentative attempt to fit their data according to Equation 27 in the form $\Delta\varrho_S(T)/\varrho_S^*(0)$ against T or $\lambda_0(T)/\lambda_0(0)$ was made by using temperature dependent values of $\varrho_0\lambda_0$. The agreement between Fuchs–Sondheimer predictions and data becomes rather good but it is only due to the use of a non-constant $\varrho_0\lambda_0$ value. Since these authors introduce $\varrho_0(T)\lambda_0(T)$ values presenting a maximum around 23 K in theoretical calculations it is obvious that this procedure will result in a closer agreement between theoretical Fuchs–Sondheimer and experimental profiles of $\Delta\varrho_S(T)/\varrho_S(0)$. Although using a temperature dependent $\varrho_0\lambda_0$ is a procedure in contradiction with the usual statement of constant $\varrho_0\lambda_0$, more important seems the incorrect choice in the value of $\varrho_0\lambda_0$ with respect to value cited in [22] which can perhaps result on surface preparation, i.e. on oxide coating on aluminium, or the presence of a disturbed surface layer. Hence concluding that the DMR mainly arises from the spatial variation of the electron distribution function and that DMR on the impurity scattering can be neglected remains questionable in the absence of information on the origin of the large $\varrho_0\lambda_0$ value deduced from the Fuchs–Sondheimer plots. It should be pointed out here that a complementary study of the film t.c.r. and of the correlated DMR $\Delta\varrho_S\Delta\beta_S$ would certainly give complementary information. The applicability of the combined Soffer–Cottey theory and the possible surface scattering origin of the DMR would be tested by fitting data using simultaneous plots of $k\Delta\varrho_S/\varrho_0$ and $k(\Delta\beta_S\Delta\varrho_S/\varrho_0\beta_0)$ against k. Unfortunately this procedure is not applicable in the temperature range investigated by Nakamichi and Kino. But it remains interesting for thin films provided that the film resistivity and t.c.r. are measured over a large temperature range (40 to 400 K for example) in order to overcome the problem of difference in surface roughness which inevitably arises when considering various films.

5. Conclusion

Combining the Cottey and the Soffer models new analytical equations are proposed to describe the r.m.s. surface roughness and the angular dependence of the t.c.r. of thin metal films. Correlated effects in the product resistivity × t.c.r. are studied. The major feature in incorporating surface roughness in calculation is to markedly diminish the overall size effect in t.c.r. with respect, in the one hand to that in the film resistivity and in the other hand to that predicted by theories involving a constant specularity parameter. Moreover a straight line relation can represent the thickness dependence in the case of moderately smooth surfaces. The deviation from Matthiessen's rule on the surface scattering is found to be conveniently represented by the variation in the product resistivity × t.c.r. and to be negligible in a large thickness range except for rough surfaces.

Attempts to fit previously published data, although nearly satisfactory, give evidence of some difficulty in interpretation that may be attributed to inaccuracies and uncertainties in measurements and in the texture of film surfaces. In particular care must be taken that films of various thicknesses certainly present differences in r.m.s. surface roughness.

A combined Soffer–Cottey model seems at first sight as convenient as the Fuchs–Sondheimer model to analyse DMR on the surface scattering in terms of theoretical expressions for the surface resistivity. However if possible a complementary investigation of the alternative DMR $\Delta\varrho_S\Delta\beta_S$ is recommended when a conclusion on the origin of DMR is required. In view of the results and to overcome some serious problems the author suggests that experimental data are taken in a large temperature range and then compared with the theoretical values related to a given specimen since a marked variation in temperature ensures a large variation in the reduced thickness.

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