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Experimental determination of the surface roughness parameter in metal films

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New approximate equations for the conductivity of metal films are derived from the theoretical predictions of the surface roughness model previously proposed to describe the effect of the rms surface roughness on the film conductivity. Comparison between exact and approximate values of the film conductivity shows good agreement in well defined thickness and roughness parameters ranges. It is found that these approximate equations are convenient tools for a systematic study of the influence of annealing temperature or condensation conditions on the film surface properties. On the basis of the present model previously published data are reinterpreted giving experimental values for the fractional change in the surface roughness due to the nucleation of a metal overlayer.

1. Introduction

It is well known [1–3] that the electrical conductivity of metals is lower in thin films than in the bulk. In the past, much experimental work [1–3] has already been interpreted in terms of the size effect theory of Sondheimer [4] which assumes that the scattering of the conduction electrons at the film surfaces can be described by a constant specularity parameter p. However, some interest [5–7] in the effect of surface roughness on the film conductivity has been revived recently. Most theoretical investigations considered the contribution of the geometrical roughness of the surface to the specularity parameter and dealt with surfaces characterized by a rms surface roughness, r, and by a gaussian form for the surface autocorrelation function [1–3, 6, 8, 9]. Another effect related to rough surfaces is the possible dependence of the specularity parameter on the angle of incidence at the surface. Some works [8–10] devoted to this study proposed a specularity parameter depending explicitly on the angle of incidence, θ.

Among the theoretical works of different authors only a few [8, 9] lead to a simple expression for the specularity parameter, p. This is particularly the case for the Soffer model [9] which includes both the influence of the rms surface roughness and the general case of oblique incidence. In effect, when the correlation length along the surface is taken to be zero, the specularity parameter p is expressed as

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

where λ_c is the wavelength associated with the carrier.

Recently, Tellier [7] offered a simple method of understanding the transport properties of thin metal films. He proposed an alternative model based on the combination of the Cottey theory [11] with the Soffer model; in this formulation the reduced film conductivity is expressed analytically in terms of the reduced surface roughness and of the reduced film thickness.

The purpose of this paper is to derive simple equations for the film conductivity in the limiting cases of small and large reduced roughness in order to perform an easy determination of the roughness parameter from experimental data obtained on films which are prepared or studied under specified experimental conditions.

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2. Simplified equations for the film conductivity

Combining the Cottey and Soffer models gives the film conductivity in the form [7]

$$\frac{\sigma_f}{\sigma_0} = \frac{3}{2}\left\{\frac{\kappa}{3}\left[\frac{1}{2}\ln\left(\frac{(\kappa+1)^2}{\kappa^2-\kappa+1}\right) + 3^{1/2}\left(\tan^{-1}\frac{2-\kappa}{3^{1/2}\kappa}+\frac{\pi}{6}\right)\right] - \frac{1}{3A}\ln(1+A)\right\} \quad (2)$$

where σ_f and σ_0 are, respectively, the film conductivity and background conductivity, and the variables κ and A are given by

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

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

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

Asymptotic expressions for the film conductivity can be derived in the limiting cases of high and low values for the parameter κ which depends on the two size parameters r and d.

In the limit of small κ expanding Equation 2 in ascending powers of κ gives the conductivity ratio in the form

$$\frac{\sigma_f}{\sigma_0} \approx \frac{\pi}{3^{1/2}}\kappa - \tfrac{3}{4}\kappa^3 + \tfrac{1}{2}\kappa^3\ln\kappa^3; \qquad \kappa < 1 \quad (5)$$

Furthermore, retaining only terms of power one the following simple relation is readily found

$$\frac{\sigma_f}{\sigma_0} \approx \frac{\pi}{3^{1/2}}\kappa; \qquad \kappa \ll 1 \quad (6)$$

which hold for very small κ.

Introducing Equations 3 and 4 into Equation 6 gives the alternative equation

$$\ln\frac{\sigma_f}{\sigma_0} \approx \frac{1}{3}\ln k - \tfrac{2}{3}\ln\frac{r}{\lambda_c} + \ln\frac{\pi}{3^{1/2}} - \tfrac{2}{3}\ln 4\pi, \qquad \kappa \ll 1 \quad (7)$$

This last equation shows that the contributions to the resistivity of the surface roughness and of the film thickness can be easily separated.

In the special case of large κ expanding Equation 2 in the power series of reciprocal κ and further neglecting terms of power higher than five one obtains

$$\frac{\sigma_f}{\sigma_0} \approx 1 - \frac{1}{8\kappa^3}; \qquad \kappa > 1 \quad (8)$$

Combining Equations 3, 4 and 8 gives the final approximate equation

$$\frac{\sigma_f}{\sigma_0} \approx 1 - \frac{2}{k}\left(\frac{\pi r}{\lambda_c}\right)^2, \qquad \left(\frac{4\pi r}{\lambda_c}\right)^2\frac{1}{k} < 1 \quad (9)$$

Note that the approximate forms of Equations 5 to 9 apply also in the more general case of two external surfaces with two different roughness parameters, r_1 and r_2, since for films with unlike surfaces it is possible to define an effective roughness expressed as [7]

$$r_{eff}^2 = \tfrac{1}{2}(r_1^2 + r_2^2) \quad (10)$$

Hence it is sufficient to replace the parameter κ by the effective parameter

$$\kappa_{eff} = \left[\frac{1}{k}\left(4\pi\frac{r_{eff}}{\lambda_c}\right)^2\right]^{-1/3} \quad (11)$$

or the surface roughness, r, by the effective roughness in the above equations to treat the case of thin metal films with unlike surfaces.

3. Discussion

In the following it would be convenient at first to achieve a graphical comparison of the theoretical predictions of the approximate Equations 6 and 8 with the predictions of the exact Equation 2.

Turning to Equation 9 it appears that in the limit of thick films and/or in the limit of small surface roughness, a plot of $\ln(1-\sigma_f/\sigma_0)$ against $\ln r/\lambda_c$ or against $\ln k$ should yield a straight line with a respective slope of 2 and -1. In effect it is easy to establish that in the case of large κ, Equation 9 can be replaced by

$$\ln\left(1-\frac{\sigma_f}{\sigma_0}\right) \approx 2\ln\frac{r}{\lambda_c} - \ln k + \ln 2\pi^2 \quad (12)$$

The variations in the function $(1-\sigma_f/\sigma_0)$ with r and k were evaluated from the exact Equation 2, k and r respectively acting as parameters (Fig. 1 and inset, respectively). In agreement with the predictions of the approximate Equation 12 we obtain straight lines with a slope

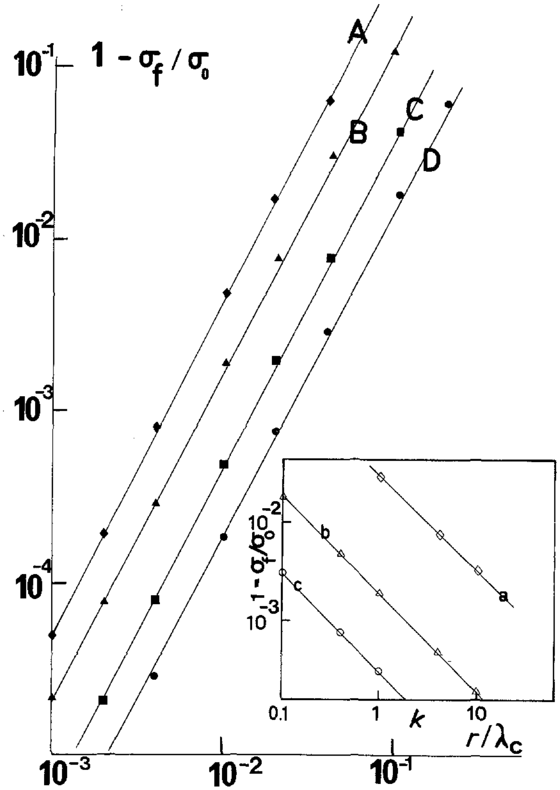

Figure 1 Plots of ln $(1 - \sigma_f/\sigma_0)$ against ln (r/λ_c), k acting as a parameter. A, B, C, D: theoretical curves for the respective k values of ♦ 0.4, ▲ 1, ■ 4 and ● 10. Inset: the plots of ln $(1 - \sigma_f/\sigma_0)$ against ln k. a, b, c: theoretical curves for the respective r/λ_c values of ◊ 0.04, △ 0.01 and ○ 0.004.

quite similar to the value calculated from Equation 12 provided that the parameter κ takes values greater than 0.9.

Moreover for thin films and/or for very rough films (i.e. for small κ) plots of the exact variations in the film conductivity with reduced thickness and reduced roughness provide further information about the range of applicability of Equation 6. As predicted by Equation 7, plots of ln σ_f/σ_0 against ln k and ln r/λ_c (Fig. 2 and inset) yield straight lines with a slope respectively equal to about 0.33 and to about -0.66 until the value of κ remains smaller than 0.25.

It would also be instructive to compare the exact values (Equation 2) with the approximate values of the reduced conductivity as given by Equations 5, 6 and 8. In Tables I and II the variations in the conductivity ratio were evaluated, r/λ_c acting as a parameter. In the limit of large κ, Table I shows that the approximate form (Equation 8) of σ_f/σ_0 accurately represents the exact form down to $k \approx 0.1$ until the reduced roughness does not exceed 0.04. In the limit of small κ, Table II shows that the deviation between the exact and approximate values of σ_f/σ_0 as given by Equations 2 and 5 respectively, remains less than 8% for $k < 0.8$ until the reduced roughness takes values greater than 0.07. When the surface roughness effect is represented by an approximate equation in the form of Equation 6 the range of applicability of this equation is reduced to $k < 0.04$ for $r/\lambda_c < 0.07$. Moreover, it appears that the k range of applicability of the approximate equations markedly depends on the value of the reduced roughness, in other words, the larger the reduced roughness the larger the k range of applicability of Equations 5 and 6 becomes.

Interest in the study of the limiting cases of the surface roughness effect is stimulated by the fact that the proposed approximate equations seem particularly adequate for a systematic investigation of the changes in the roughness parameter r on annealing or on deposition conditions. Effectively, many experiments [1, 3, 12–15] have revealed that at smaller annealing temperatures a surface reordering phenomenon

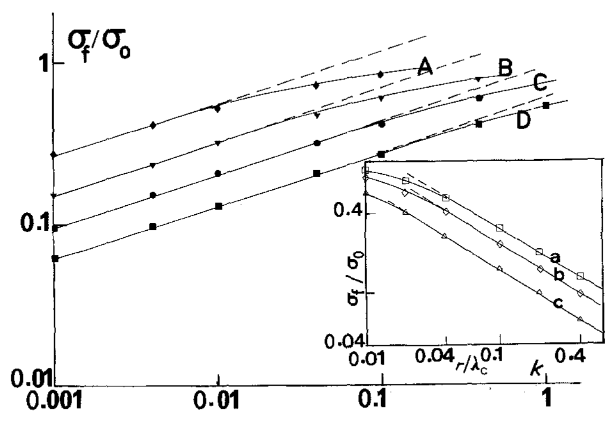

Figure 2 Plots of ln σ_f/σ_0 against ln k, r/λ_c acting as a parameter. A, B, C, D: theoretical curves for the respective r/λ_c values of ♦ 0.04, ▼ 0.1, ● 0.2 and ■ 0.4. Inset: the plots of ln σ_f/σ_0 against ln r/λ_c. a, b, c: theoretical curves for the respective k values of □ 0.01, ○ 0.004 and △ 0.001.

TABLE I Comparison of the values of the conductivity ratio σ_f/σ_0 evaluated from Equations 2 and 8

k	Exact values (Equation 2)			Approximate values (Equation 8)		
	$r/\lambda_c = 0.04$	$r/\lambda_c = 0.02$	$r/\lambda_c = 0.01$	$r/\lambda_c = 0.04$	$r/\lambda_c = 0.02$	$r/\lambda_c = 0.01$
0.1	0.823 39	0.935 47	0.981 36	0.684 17	0.921 04	0.980 26
0.2	0.889 13	0.964 61	0.990 42	0.842 09	0.960 52	0.990 13
0.4	0.935 47	0.981 36	0.995 14	0.921 04	0.980 26	0.995 07
1	0.971 11	0.992 29	0.998 04	0.968 42	0.992 10	0.998 03
2	0.984 92	0.996 10	0.999 02	0.984 21	0.996 05	0.999 01
4	0.992 29	0.998 04	0.999 51	0.992 10	0.998 03	0.999 51
10	0.996 87	0.999 21	0.999 80	0.996 84	0.999 21	0.999 80

generally occurs, affecting the surface roughness parameter. For example, since for films annealed at temperatures greater than 200 K we are generally concerned with relatively large reduced thicknesses, the approximate Equation 12 seems convenient for following the progressive reordering of the upper surface of films in the range of annealing temperatures in which the volume defects (vacancies, dislocations or grain boundaries) remain unannealed. In effect, for a reference temperature $T_0 \simeq 200$ K, Equation 12 predicts that a plot of the film conductivity in the form $\ln [1 - \sigma_f/\sigma_0]$ against $\ln r/\lambda_c$ should yield a straight line with a slope equal to 2. But care must be taken that the use of Equation 12 requires corrections on the experimentally measured values of the film conductivity, on the bulk conductivity and on the bulk mean free path, to take into account the variations in these parameters with annealing temperature.

Quartz crystal microbalances [16] are now commonly used to monitor the film thickness during vacuum evaporation. Hence for films deposited at low temperatures, Equations 6 and 7 can constitute convenient ways to study the effect of deposition conditions on the geometrical properties of the upper surface provided that the deposited films do not exhibit a grained structure with a mean grain size which varies markedly with the film thickness or with deposition conditions [17]. For example, in the low temperature range and for a given film thickness, Equation 7 predicts linear variations of $\ln \sigma_f/\sigma_0$ with increasing $\ln r/\lambda_c$, characterized by a negative slope of -0.666. Hence this approximate equation allows us to estimate easily the surface roughening of a film by various treatments.

Interest in the problem of approximate expressions of film conductivity is also revived by inspection of Equation 7 which reveals that the physical parameter, r, can be easily extracted, with a reasonable accuracy and suggests a convenient procedure for a systematic study of the changes in the resistivity of annealed films during the nucleation of a metal overlayer [18–23]. At this point it is essential to compare the theoretical predictions of the present approximate model with experimental data. But the comparison is significant only if the bulk properties of all films are nearly the same so that variations in the film resistivity during successive overlayer nucleations can be attributed without ambiguity to changes in the upper surface properties. In view of this difficulty a comprehensive interpretation of the surface roughness effect in terms of the proposed model can be, to our knowledge, undertaken only with data on expitaxial silver films previously reported by Berman and Juretschke [21]. To identify detailed aspects of surface scattering, these authors controlled the surface roughness of the upper surface by superimposing, at the liquid nitrogen temperature T_N, a very thin silver overlayer on a silver base film thoroughly annealed below room temperature. The new sample was then submitted to a specified temperature cycle and the procedure was repeated to give a film of total thickness of about 100 nm. Resistivity measurements revealed that the overlayer was fully annealed at a temperature, T_S, of about 250 K. At temperatures in the range $T_N < T < T_S$ the increase of the resistivity was interpreted in terms of the effect of a very rough overlayer since for a fully annealed overlayer a slight decrease of the film resistivity was observed, as expected, for a final film whose thickness has increased slightly.

A tentative evaluation of the fractional change, $\Delta r/r$, of the surface roughness on annealing in terms of the approximate Equation 6 can be made even if, unfortunately, in the temperature range $T \geqslant T_N$ the reduced film

TABLE II Comparison of the values of the conductivity ratio σ_f/σ_0 evaluated from Equations 2, 5 and 6. Values omitted in this table correspond to physically unreasonable values (i.e. $\sigma_f/\sigma_0 > 1$)

k	Exact values (Equation 2)			Approximate values (Equation 5)			Approximate values (Equation 6)		
	$r/\lambda_c = 0.1$	$r/\lambda_c = 0.2$	$r/\lambda_c = 0.4$	$r/\lambda_c = 0.1$	$r/\lambda_c = 0.2$	$r/\lambda_c = 0.4$	$r/\lambda_c = 0.1$	$r/\lambda_c = 0.2$	$r/\lambda_c = 0.4$
0.001	0.152 95	0.097 31	0.061 58	0.152 95	0.097 31	0.061 58	0.155 76	0.098 12	0.061 81
0.002	0.191 07	0.122 11	0.077 45	0.191 07	0.122 11	0.077 45	0.196 24	0.123 63	0.077 88
0.006	0.269 59	0.174 28	0.111 15	0.269 59	0.174 28	0.111 15	0.283 03	0.178 30	0.112 32
0.01	0.314 78	0.205 10	0.131 32	0.314 79	0.205 10	0.131 32	0.335 57	0.211 40	0.133 17
0.02	0.385 59	0.254 85	0.164 36	0.385 63	0.254 86	0.164 36	0.422 79	0.266 34	0.167 78
0.06	0.518 86	0.354 87	0.233 03	0.519 15	0.354 89	0.233 03	0.609 77	0.384 13	0.241 99
0.1	0.587 31	0.410 70	0.272 99	0.588 10	0.410 75	0.272 99	0.722 96	0.455 44	0.286 91
0.2	0.681 95	0.495 21	0.336 38	0.685 04	0.495 41	0.336 39	0.910 88	0.573 82	0.361 48
0.6	0.818 94	0.642 79	0.459 01	0.844 90	0.644 54	0.459 13	–	0.827 59	0.521 35
1	0.870 06	0.711 79	0.524 22	0.937 97	0.716 58	0.524 53	–	0.981 21	0.618 12

TABLE III Reduced surface roughness and fractional changes in surface roughness using the exact and the approximate models. The subscripts R and A are, respectively, related to parameters after deposition of a thin rough overlayer and to parameters after full anneal. At 100 K the bulk parameters ϱ_0 and λ_0 are assumed to be, respectively, equal to about 0.428 $\mu\Omega$ cm and 210 nm [21]

	Total film thickness (nm)		
	29	69	110
σ_f/σ_{0R}	$\simeq 0.264$	$\simeq 0.428$	$\simeq 0.507$
r/λ_{cR} (Equation 2)	0.45	0.34	0.31
r/λ_{cR} (Equation 6)	0.536	0.40	0.389
σ_f/σ_{0A}	$\simeq 0.386$	$\simeq 0.581$	$\simeq 0.732$
r/λ_{cA} (Equation 2)	0.26	0.185	0.135
r/λ_{cA} (Equation 6)	0.301	0.252	0.224
$\frac{\Delta r}{r_R}$ (Equation 2)	46%	46%	56%
$\frac{\Delta r}{r_R}$ (Equation 6)	44%	37%	42%

thickness takes values which lie in the limit of validity of the present equation. In order to estimate the fractional changes in the reduced roughness due to annealing we have analysed the data [21] at 100 K using the exact Equation 2 and the approximate Equation 6. The results of this procedure are summarized in Table III for un-annealed and annealed overlayers. Table III also lists the bulk mean free path and the bulk resistivity in agreement with the values previously reported by Berman and Jurestschke [21]. From a practical point of view, data interpreted in terms of the exact and the approximate models lead to quite similar values of the fractional decrease in the reduced roughness with thermal ageing. It must be pointed out that for films of 69 and 110 nm the deposition of an overlayer on a thoroughly annealed base film induces an increase of the surface roughness to about the same value. Moreover, it seems that the relative decreases in the reduced roughness due to annealing do not depend markedly on the base film thickness when the data are treated according to Equations 2 and 6. We thus observe a similarity of the predictions of the exact and the approximate models.

4. Conclusions

Simple approximate equations are proposed to describe the surface roughness and the thickness dependence of the conductivity of metal films. The conditions of validity for a good agreement between the approximate values of the film conductivity as deduced from the present model and the exact values derived by combining the Soffer and Cottey [9, 11] models, are determined precisely.

The above discussion shows that the approximate equations can be convenient tools for evaluating the variations in the surface roughness induced by thermal ageing, by making modifications to deposition conditions or by the nucleation of a metal overlayer.

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