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Study of the size effects in the electrical resistivity of ultrathin (<Cu 40 nm) films in the framework of the statistical conduction models

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ABSTRACT

In their pioneering experimental work, Liu et al. have given the data related to the in situ sheet resistance measurements of polycrystalline ultrathin Cu films, where the resistivity ρ, was determined as a function of film thickness d.

The aim of this paper is to show that the size effects in polycrystalline ultrathin Cu films can be easily reinterpreted by using a simple analytical expression of the electrical conductivity, earlier proposed in the framework of the multidimensional conduction models. The electronic transport parameters obtained in this study are in good agreement with our previous theoretical works. For this purpose, the study given by the authors which has been interpreted by using the Namba's model is reconsidered.

1. Introduction

The foundations of the modern electron theory of metals [1] were laid at the beginning, when Drude has postulated the existence of gas of free electrons, in order to explain the conducting properties of metals which cannot explain why the conduction electrons do not contribute to the specific heat of metals. This question was solved by Pauli and Somerfield by applying the Fermi-Dirac statistics. On the behavior of the electrical resistivity of thin films, several scattering terms as a result of defects, surfaces, grain boundary and impurity scattering, as well as morphological defects, most studies have considered only the surface scattering and the grain boundary. The transport mechanisms and in turn the cause of resistance are of fundamental importance. Various models especially for thin films exist to understand the contribution of different scattering mechanisms.

The electrical conductivity of thin metal films was studied by Fuchs and Sondheimer [2] and their theories were developed to explain the fact that in thin films of alkali metals, the resistivity is always higher than in bulk materials, and increases rapidly as the thickness decreases. The electrical conductivity of sufficient thin metal films is less than that of the bulk material. The F–S analysis is based on the following simplifying assumptions:

- The energy of surfaces is spherical (as in the quasi free electron model) so that the relaxation time and consequently the electron mean free path can be regarded as constant over the Fermi-surface.
- The specularity parameter P is independent of the electron energy and of the angle of incidence to the surface.
- The Boltzmann's equation is written for the particular case where a single parameter P can describe the surface scattering. Indeed this assumption does not take into account the different kinds of surface scattering and especially, as pointed out by Greene [3,4], the scattering at the crystal surface by localized surface charges.

Some authors [5,6] have presented some sophisticated models in order to solve the problem of surface scattering when one of the assumptions mentioned above is not fulfilled. These models lead generally to complicated equations which do not allow simple single interpretation of experimental data. Namba [7] proposed a model which includes a surface roughness in addition to surface and interface scattering when calculating metal film conductivity. The film thickness in this model is assumed to have undulations of an amplitude H. However, it must be kept in mind that the Parott [5] model is not entirely devoted to the study of the influence of the surface structure on the specularity, but to the effect of non-spherical energy surfaces on the film conductivity.

On one hand, Mayadas and Shatzkes [8] have proposed a unidimensional model for the total film conductivity by taking into account the effect of the grain boundaries, with the simplifying assumptions that only the grain boundaries perpendicular to the electric field induce electronic scattering. And, on the other hand,

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they suggested that it might be hoped that an effective mean free path (m.f.p.) could be defined for a polycrystalline films, but no theoretical basis has been proposed for this point of view. Since the publication of this model, some papers related to electrical conductivity have used M–S equation or M–S asymptotic equations for interpreting the experimental data.

The structure of thin metallic films has provided evidence for the existence of fine-grained structures when the films were deposited under well determined conditions. This fact necessitates the study of a theoretical model for calculating the resistivity of fine-grained metal films caused by grain boundaries scattering processes.

Starting from the similar assumption made by Mayadas and Shatzkes [8] and taking into account the above features in the three dimensional models [9,10], it is assumed that the grain boundaries in polycrystalline metal films can be represented by three arrays of mutually perpendicular planar potentials with rough surfaces oriented perpendicular to the *X*-, *Y*- and *Z*-axes, respectively. The current is due to electron which has been transmitted through a large number of grain boundaries and it is assumed that *T* is the fraction of electrons which are specularly transmitted through the grain boundaries whilst for the remainder the free path is as usual terminated by collision at the boundary.

The thickness dependence of the resistivity for Cu films deposited by ion beam deposition has been studied by Lim et al. [11] and the electrical transport parameters in metallic films was evaluated using Fuchs–Sondheimer and Mayadas–Shatzkes models.

In their pioneering experimental work, Liu et al. [12] have given the data related to the in situ sheet resistance measurements of polycrystalline Cu films, where the Cu film resistivity ρ was determined as a function of film thickness d. The Cu films were deposited at room temperature at a rate of 2.56 ± 0.2 nm/min, from a radiation heated Cu foil subject to electron bombardment. The size effects in the electrical resistivity of ultrathin Cu films are reexamined in this paper.

2. Theoretical aspects

2.1. Namba's model

Sophisticated models for the surface roughness models namely Ziman [1], Soffer [13] and Namba [7] methods have been proposed. The simplest method proposed to determine the effect of the geometrical roughness of the surface on film conductivity is that proposed by Namba [7]. Contrary to Ziman [1] and Soffer [13] formulations which are based on optical arguments, Namba's [7] model deals only with a geometrical surface model and is not concerned with a theoretical formulations of the specularity parameter *P*.

Considering the forms exhibited by thin film cross-sections, Namba [7] has assumed that in the thin film resistivity calculations the small bumps will be taken into account in terms of the F–S specularity parameter whereas the large undulations will be considered as local charges in film thickness. In these conditions the problem is reduced to determining the film thickness distribution $d(x)$ due to undulations. Namba [7] assumes that the film thickness can be represented by

$$d(x) = d + H\sin\left(\frac{2\pi}{s}x\right) \tag{1}$$

where *d* is the average thickness, *s* the undulation length, and *H* the amplitude of the sinusoidal undulation; the electrical conductivity in this case becomes:

$$\sigma[d(x)] = \sigma_\infty\left[1 - \frac{3\lambda_0}{2\mathrm{d}(x)}(1 - P(x))\right] \times \int_0^\infty \left[\frac{1}{t^3} - \frac{1}{t^5}\right]\frac{1 - e^{\left(-(d(x)/\lambda_0)t\right)}}{1 - P(x)e^{\left(-(d(x)/\lambda_0)t\right)}}\,dt \tag{2}$$

where $t = 1/\cos\theta$ and θ is incidence angle of the electrons at the surface.

2.2. Statistical model

Previous works [10] have shown that a unique equation can be used for describing the electrical conductivity of thin metal films, by taking into account simultaneously the scattering due to phonons, external surfaces and grain boundaries. It has been established [10] that whatever the film structure and the roughness of the film surface, the electrical conductivity σ_f, is given by [10,14]:

$$\sigma_f = \sigma_0 C(\mu, \nu) \tag{3}$$

where

$$C(\mu, \nu) = \frac{3}{2b}\left[a - \frac{1}{2} + (1 - a^2)\ \ln\left(1 + \frac{1}{a}\right)\right] \tag{4}$$

With

$$\mu = \frac{K}{2}\left[\frac{1 + p}{1 - p}\right] \tag{5}$$

$$\nu = \frac{D_g}{2\lambda_0}\left[\frac{1 + T}{1 - T}\right] \tag{6}$$

$$b = \frac{1}{\mu} + \frac{c_1}{\nu} \tag{7}$$

$$a = \frac{1}{b}\left(1 + \frac{c_2}{\nu}\right) \tag{8}$$

$$c_1 = 1 - c \quad \text{for polycristalline films} \tag{9}$$

$$c_1 = -c \quad \text{for monocristalline or columnar film} \tag{10}$$

$$c = \frac{4}{\pi} \tag{11}$$

σ_0 is the electrical conductivity of the bulk material, *d* the film thickness, λ_0 the bulk mean free path, $K = d/\lambda_0$ the reduced thickness, *P* the specular reflection coefficient at the film surface, D_g the grain boundary size and *T* is the specular transmission coefficient at the grain boundary which gives the fraction of electrons whose velocity in the electric field direction is not altered by the grain boundary, whereas the remainder of the electrons are diffusely scattered and do not contribute to the current [9,10]. Assuming that the probability that electrons have travelled a distance without being scattered boundary, is given by an exponential law [10]; a mean free path can be ascribed to the tree-dimensional array of scatterers. Moreover a parameter, ν has been introduced, known as grain boundary parameter and defined by Eq. (6). The statistical models can be easily used for interpreting the size effects in thin metallic films, double layers and thin wires. New formulations of both the electrical resistivity and its coefficient of temperature (t.c.r.) have been recently proposed [15].

The asymptotic expression for the reduced conductivity has been shown [14], as follows:

$$\frac{\rho_f}{\rho_0} \approx ba + c_2 b \quad a \gg 0.1 \tag{12}$$

With

$$c_2 = 0.375 \tag{13}$$

The resistivity at infinite thickness ρ_∞ is

$$\rho_\infty = \rho_0\left(1 + \frac{c^2 + c_1 c_2}{\nu}\right) \tag{14}$$

The electrical resistivity ρ_f can then be expressed by using that obtained at infinite thickness ρ_∞ as:

$$\rho_f = \rho_\infty + \rho_0\frac{c_2}{\mu} \tag{15}$$

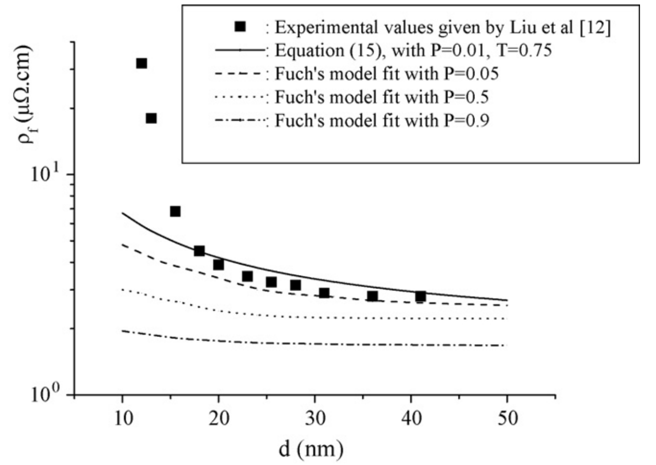

Fig. 1. Variations of the electrical resistivity ρ_f with thickness d, from Eq. (15) with ($P = 0.01$, $T = 0.75$), Fuchs' model fits with P (0.05, 0.5 and 0.9) and experimental data.

3. Results and discussion

In the case of thermally evaporate ultrathin Cu films with thickness d between 10 and 40 nm onto 500 nm thick SiO_2 on Si(1 0 0) substrates, Liu et al. [12] have used Namba's model [7] for interpreting their experiments.

Figs. 1–3 represent the dependence of the electrical resistivity, related to experimental data, the computed values obtained from the F–S, M–S, Namba's models given by Liu et al. [12] and those calculated from Eq. (15):

- Fig. 1 shows that the results obtained by using Eq. (15) with $P = 0.01$, $T = 0.75$, which describe the experimental data are more precise than those given by the authors [12] in the case of the F–S model fitted for three values of P (0.05, 0.5 and 0.9).
- Fig. 2 represents the experimental data, the fits obtained in the Namba's model with $P = 0.05$ and two values of H ($H = 10.3$ and $H = 3.5$ nm) and the calculated values obtained from Eq. (15), with $P = 0.01$, $T = 0.75$. In this respect, only the Namba's model for the value $H = 10.3$ nm gives the best fit with the experimental data in particular case for the lowest thickness.
- Fig. 3 gives the experimental data, the calculated values obtained from Eq. (15), with $P = 0.01$, $T = 0.75$ and the fit obtained in the M–S model with $P = 0.05$, $R = 0.24$. It must be also noted that the best fit given by the authors [12] is obtained by assuming that a reduced grain size is equal to $10d/3\lambda_0$; this hypothesis has also been considered in our calculations. In one way the calculated resistivity given by the authors [12] matches well the experiments only for $d - 20$–40 nm, however the calculated resistivity is lower compared with the experimental data, and in another way it has been established in a previous work [9] that the M–S model is unidimensional and therefore no precise physical significance can be attached to the so-called electronic reflection coefficient R.

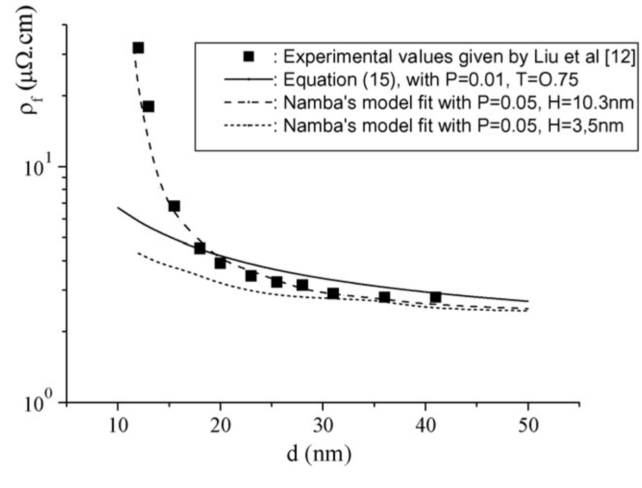

Fig. 2. Variations of the electrical resistivity ρ_f, with thickness d, from Eq. (15) with ($P = 0.01$, $T = 0.75$), Namba's model with ($P = 0.05$, $H = 10.3$ nm), ($P = 0.05$, $H = 3.5$ nm) and experimental data.

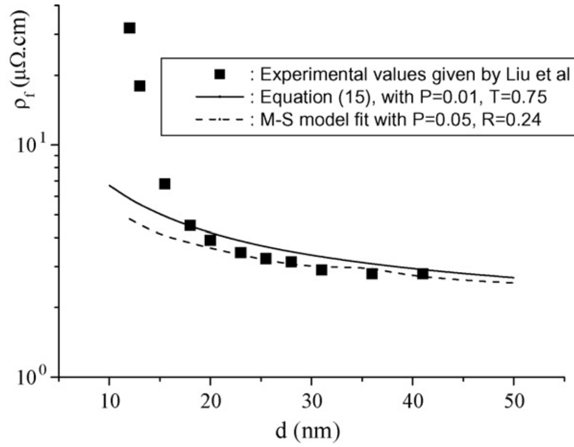

Fig. 3. Variations of the electrical resistivity ρ_f, with thickness d, from Eq. (15) with ($P = 0.01$, $T = 0.75$), M–S model with ($P = 0.05$, $R = 0.24$) and experimental data.

It has been early established [9,10,14] that in thin polycrystalline films in which three types of electrons scatterings, i.e. background scattering, grain boundary and external surface scattering, are simultaneously operative, a three dimensional models are very similar to the M–S model [10] and then can be regarded as an alternative algebraic formulation for the complicated expression obtained by Mayadas and Shatzkes.

In this work we show that the size effects in ultrathin (<Cu 40 nm) films can be easily analyzed in terms of:

- Extended Cottey's model [10], in which a mean free path is associated with scattering; an "external surface" parameter μ, given by Eq. (5).
- Statistical models, where the effects of the grain boundaries are described by the grain parameter ν, given by Eq. (6).

A new plot of the experimental data [12] is represented in Figs. 4 and 5. It appears clearly that these figures are in good agreement with the asymptotic equation (15) in the whole range of the experimental thickness, except at the lowest thickness (two experimental data, only). Furthermore, it must be noted that the author's experiments have shown that the electrical resistivity dropped from 32.34 ± 0.50 to 3.09 ± 0.03 μΩ cm, when the thickness increased from 11.5 to 15.3 nm. This behavior can interpret accordingly the author's observation and suggest that the Cu films grown on SiO_2 substrates are discontinuous at the lowest thickness.

By taking into account the values of the bulk resistivity and that the electronic mean free path used by the authors ($\rho_0 = 1.69$ μΩ cm and $\lambda_0 = 39$ nm), the slope of the linear film resistivity, ρ_f, against the reciprocal thickness (Fig. 4), as derived from Eqs. (5) and (15), gives the value of the effective reflection coefficient P; the numerical data obtained is reported in Table 1, which is in substantial agreement over that given by the authors [11,12].

Fig. 5 represents the linear variation of the product $\rho_f d$ against d, which gives the value of ρ_∞. Eqs. (6) and (14) are used for calculating the specular electronic transmission coefficient T (Table 1). The

Table 1
Electronic transport parameters in the different models.

	Specular reflection coefficient, *P*				Reflection/ transmission coefficient at grain boudaries		Roughness
	F–S model	M–S model	Namba's model	Statistical model	*R*	*T*	Namba's model
From Ref. [11]	0.00	0.00			0.4		
From Ref. [12]	0.05	0.05	0.05		0.24		10.3 nm
From Eq. (15)				0.01		0.75	

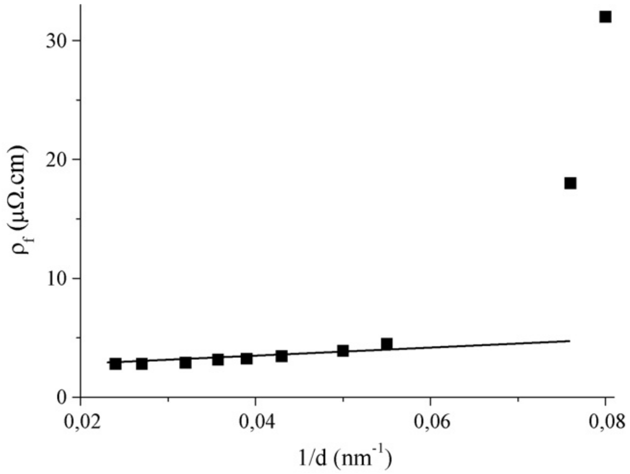

Fig. 4. Variations of the electrical resistivity ρ_f with reciprocal thickness d^{-1}.

obtained value of T is in very good agreement with that deduced from the relation between R (equal to 0.24) in the M–S model and the specular reflection coefficient T (near 0.75) in the statistical models, which has been proven [10] by the following equation:

$$T = \frac{2 - 2.9R}{2 - 1.1R} \tag{16}$$

The electrical parameters obtained in this work, the computed values given by Liu et al. [12] and those deduced from Lim et al. [11] are summarized in Table 1. The reflection coefficient R of the thermally evaporated Cu films given by Liu et al. [12] is equal to 0.24 and that related to the Cu films deposited by ion beam deposition (I.B.D.) calculated by Lim et al. is equal to 0.4. The study conducted by the authors, makes us giving the following remarks:

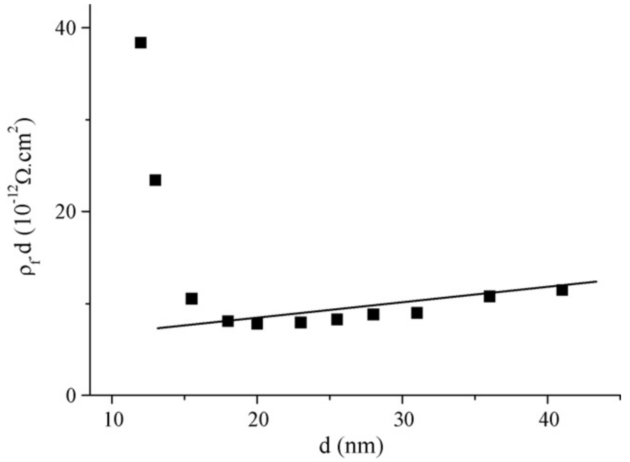

Fig. 5. Variations of the product $\rho_f d$ with the thickness d.

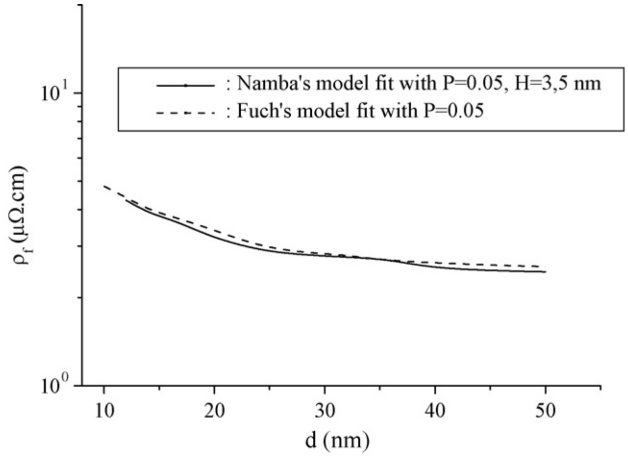

Fig. 6. Comparison between the computed fits given by the Namba's model with (P = 0.05, H = 3.5 nm) and the F–S model for P = 0.05.

- The high value of the roughness 10.3 nm, which gives the best fit using Namba's model, this seems to us is to high in relation to the considered lowest thickness (d < 15 nm); at the mean time the AFM image used by the authors revealed that the roughness is 2.6 ± 0.50 nm, which is lower than the above computed value.
- The computed fit obtained by the use of Namba's model at the roughness value equal to 3.5 nm, and the fit calculated from the F–S model for P = 0.05, as represented in Fig. 6; leads to a close agreement between the two models.
- Using Namba's model; the authors are neglecting the effect of the grain boundaries. This view matches the precedent remark.
- It is well known [9,10,14] that as the film thickness d decreases and approaches the m.f.p., the film resistivity increases due to an increased relative contribution from the external surface and grain boundaries scattering. We believe that these results can be easily reinterpreted by using Eq. (15) deduced in the framework of the multidimensional models.

4. Conclusion

This study concluded that the experimental data related to the thermally evaporated ultrathin (<Cu 40 nm) can be easily reexamined by the use of Eq. (15) and the sophisticated model of Namba and the computed fittings given by the authors are not necessary for interpreting the experiments. The effects of the three types of scatterings are simultaneously taken into account and the value of the deduced specular reflection coefficient is very near from that given by the authors. The effect of the grain boundary scattering is represented by the transmission coefficient T which its value matches our previous works.

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