Vol. A54 (1978) ACTA PHYSICA POLONICA No. 1

ELECTRICAL CONDUCTIVITY OF THIN METAL FILMS. SIZE EFFECTS

By F. Warkusz

Institute of Physics, Wrocław Technical University*

(Received November 15, 1977)

An expression for the resistivity of metal films, which takes into account the thickness of the film, the coefficient of surface scattering, the grain diameter, and the coefficient of electron travelling through the grain boundary, is derived.

1. Introduction

The electrical conductivity of thin metal films is less than that of the bulk material. This is due to the reflection of electrons from bounding areas and grain boundary, as well as to the imperfections of the crystalline lattice whose density increases in thin films.

The Fuchs-Sondheimer [1, 2] model takes into account surface scattering (expressed in terms of the coefficient of specular reflection on external surfaces (p)), the mean free path of the bulk material (λ_B), and film thickness (a). Fuchs theory was developed by Cottey [3], who has introduced a layer model for smooth films, and by Moraga and Vilche [4], who have involved the same layer model for inhomogeneous films.

In the literature, many experiments are reported which relate conductivity to the crystalline grain size [5–10]. Wedler and Wissmann [9, 11] have derived an approximate expression for the electrical resistivity of thin polycrystalline metal films. The expression also includes the grain-boundary scattering effect. The correlation between the Mayadas--Shatzkes [8] and the Wissmann-Wedler models was studied by Thieme and Kirstein [12]. Mola and Heras [13, 14] as well as Tellier and Tosser [15], involving the well-known Mayadas-Shatzkes model, have derived approximate equations for the thickness dependence of resistivity, which include both grain-boundary scattering and surface scattering.

* Address: Instytut Fizyki, Politechnika Wrocławska, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland.

2. *Theory*

In the present paper, the electrical conductivity of thin metal films is calculated, taking into account both surface scattering and grain-boundary scattering effects, and introducing a layer model. The calculations are performed in terms of the free electron model. Electron scattering at the external surface and grain-boundary scattering are assumed to take place independently.

Let us consider three particular cases.

(*i*) Continuous metal films with determined thickness are considered. The electrons approach the upper surface of the film at angle θ, $(0 \leqslant \theta < \pi/2)$, while electrons which approach the lower surface of the film have $\pi/2 < \theta \leqslant \pi$ (Fig. 1). The coefficient of the specular reflection from a single plane is p, $(0 \leqslant p \leqslant 1)$. If the electrons move at angle θ

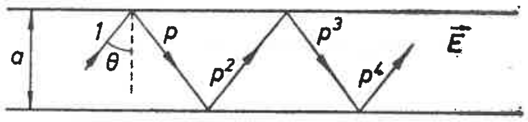

Fig. 1 Model of a continuous film

and are scattered from n surface at a film thickness a, then the travelling distance l between layers becomes $l = na/|\cos\theta|$, and the probability of specular reflection at the travelling distance l may be expressed as follows

$$P = p^n, \quad \text{where} \quad n = \frac{l|\cos\theta|}{a}. \tag{1}$$

It is assumed (after Cottey) that P varies proportionally to the travelling distance l, we obtain

$$P = \exp(-l/\lambda_a), \tag{2}$$

where λ_a is the mean free path associated with the influence of conduction electrons of film surfaces. Combining Eq. (2) and Eq. (1) we get

$$\lambda_a(\theta) = \frac{a}{|\cos\theta| \ln(1/p)}. \tag{3}$$

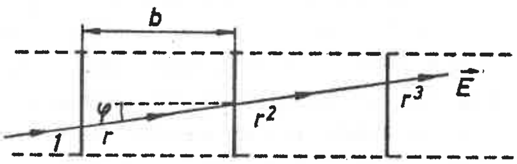

Fig. 2. Model of the grain structure

(*ii*) In the present paper the grain-boundary scattering phenomenon is taken into account and a grain-boundary scattering model introduced. The grains are assumed to have equal diameters, and grain boundaries are assumed perpendicular to the film planes (Fig. 2). The electrons strike the grain boundary at angle $\varphi = \pi/2 - \theta$, $(0 < \theta < \pi)$.

Let the probability of electron travelling through a single grain boundary be r, $(0 \leqslant r \leqslant 1)$, and the probability of travelling through m grain boundaries be r^m. The probability of electron travelling at distance l (without changing the travelling directions) becomes

$$P = r^m = r^{\frac{l \sin \theta}{b}}, \tag{4}$$

where $l = m\,b/\sin\theta$ and b is grain diameter.

It is also assumed [16] that $P = \exp(-l/\lambda_b(\theta))$, where $\lambda_b(\theta)$ denotes the mean free path associated with the influence of conduction electrons on the grain boundary and takes the form

$$\lambda_b(\theta) = \frac{b}{\sin\theta \ln(1/r)}. \tag{5}$$

(*iii*) Considering (*i*) and (*ii*), as well as the independence of the electron mean free paths λ_a, λ_b and λ_B, the electron scattering model for a real film is developed. The model of a real film is shown in Fig. 3. The beam of conduction electrons passes through the

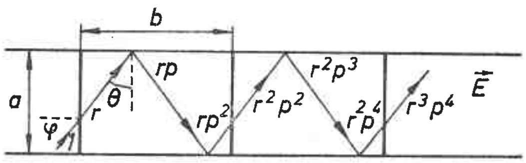

Fig. 3. Model of the real film structure

grain boundary, is scattered by the external surfaces, and varies in intensity with varying coefficients r and p. If the electrons at the travelling distance l penetrate m grain walls and are scattered by n surfaces, then the probability of such an electron travelling distance l is

$$P(r, p) = r^m p^n = e^{-\frac{l}{\lambda_b(\theta)}}\, e^{-\frac{l}{\lambda_a(\theta)}}. \tag{6}$$

Let the bulk scattering mean free path be λ_B, and assume λ_B independent of direction and film thickness, we can write the mean free path $\lambda(\theta)$ for the resultant scattering either as

$$\frac{1}{\lambda(\theta)} = \frac{1}{\lambda_B} + \frac{1}{\lambda_a(\theta)} + \frac{1}{\lambda_b(\theta)}. \tag{7}$$

The formula for the electrical conductivity [1, 3] of the film becomes

$$\sigma = \frac{I}{E} = \frac{2\pi e^2 P_F^2}{h^3} \int_0^{\pi} \sin^3 \theta \lambda(\theta) d\theta, \tag{8}$$

where P_F is the Fermi momentum of the electron, and h denotes the Planck constant, $\lambda(\theta)$ as in Eq. (7).

Combining the value of $\lambda(\theta)$ from Eqs (7), (3) and (5) with Eq. (8) we obtain

$$\frac{\sigma_F}{\sigma_B} = \tfrac{3}{4}\mu\nu \int_0^{\pi} \frac{\sin^3\theta}{\mu\nu+\nu|\cos\theta|+\mu\sin\theta}\, d\theta, \tag{9}$$

where

$$\mu = \frac{a}{\lambda_B \ln(1/p)}, \tag{10}$$

$$\nu = \frac{b}{\lambda_B \ln(1/r)}, \tag{11}$$

and σ_B is the bulk conductivity. Eq. (9) has been transformed into the integral of a rational function. Then the integrand has been represented as a sum of fractions.

The solution of the integral (9) includes the following expressions:

$$\begin{aligned}\frac{\sigma_F}{\sigma_B} &= W_1(\mu,\nu) \quad \text{for } \mu \neq 1,\ \Delta > 0 \text{ (If } \mu \neq 1, \text{ then } \Delta \neq 1)\\ &= W_2(\mu,\nu) \quad \text{for } \mu = 1,\ \Delta > 0 \text{ (If } \mu = 1, \text{ then } \Delta = 1)\\ &= W_3(\mu,\nu) \quad \text{for } \Delta = 0\\ &= W_4(\mu,\nu) \quad \text{for } \Delta < 0,\end{aligned}$$

where

$$\Delta = \mu^2 - \mu^2\nu^2 + \nu^2,$$

$$W_1(\mu,\nu) = 24\mu\left\{A\ln\left(\frac{\nu(\mu-1)}{\mu-\sqrt{\Delta}}+1\right) + B\ln\left(\frac{\nu(\mu-1)}{\mu+\sqrt{\Delta}}+1\right) + \tfrac{3}{16}C + \frac{3\pi+8}{32}D + \tfrac{1}{4}E + \frac{\pi+2}{8}F + G\ln\sqrt{2} + \frac{\pi}{4}H\right\}, \tag{12}$$

$$W_2(\mu=1,\nu) = \frac{12\nu}{(\nu^2+1)^2}\left\{\frac{\nu^3}{\nu^2+1}\left(\ln\frac{\nu\sqrt{2}}{1+\nu} - \frac{\nu\pi}{4}\right) + \tfrac{1}{32}(5\pi\nu^2+8\nu^2+\pi+2\nu^3-6\nu)\right\}, \tag{13}$$

$$W_3(\mu,\nu) = \frac{9}{\nu^2\mu}\ln\frac{\mu\sqrt{2}}{\mu+\nu\mu-\nu} - \frac{3(\mu-1)}{\nu(\mu+\nu\mu-\nu)} + \frac{3(3\mu\pi+4\mu-3\pi)}{4\nu^3(\mu-1)} - \frac{3(3\pi+8)}{8\nu} + \frac{3(2-\mu)}{2\nu^2(\mu-1)} - \frac{9}{4\mu}, \tag{14}$$

$$W_4(\mu,\nu) = 24\mu\left\{\frac{A'}{2}\ln\frac{2\mu(\nu+1)}{\nu(\mu+1)} + \frac{B'\nu - A'\mu}{\sqrt{-\Delta}}\arcsin\sqrt{\frac{-\Delta}{2\nu\mu(\mu+1)(\nu+1)}} + \tfrac{3}{16}C + \frac{3\pi+8}{32}D + \tfrac{1}{4}E + \frac{\pi+2}{8}F + G\ln\sqrt{2} + \frac{\pi}{4}H\right\}, \tag{15}$$

where

$$A = \frac{-\nu^4(\mu-1)^3(\mu-\sqrt{\Delta})^3}{2\sqrt{\Delta}\{(\mu-\sqrt{\Delta})^2+\nu^2(\mu-1)^2\}^3}, \quad B = \frac{\nu^4(\mu-1)^3(\mu+\sqrt{\Delta})^3}{2\sqrt{\Delta}\{(\mu+\sqrt{\Delta})^2+\nu^2(\mu-1)^2\}^3},$$

$$C = \frac{-\nu^2}{2(\nu^2+\mu^2)}, \quad D = \frac{-\mu\nu}{2(\nu^2+\mu^2)},$$

$$E = \frac{\nu^2\{\nu^2(\mu+1)-\mu^2(\mu-1)\}}{4(\nu^2+\mu^2)^2}, \quad F = \frac{\mu\nu\{\nu^2(\mu+1)+\mu^2\}}{2(\nu^2+\mu^2)^2},$$

$$G = \frac{\nu^4\{\mu^2(3\mu^2+1)-\nu^2(\mu^2-1)\}}{8(\nu^2+\mu^2)^3}, \quad H = \frac{\nu^2\mu\{(\nu^2+\mu^2)(\mu-1)^2-4\nu^2\mu^2\}}{8(\nu^2+\mu^2)^3},$$

$$A' = \frac{-\nu^4\{\mu^2(3\mu^2+1)-\nu^2(\mu^2-1)\}}{8(\nu^2+\mu^2)^3}, \quad B' = \frac{-\nu^3\mu(\mu+1)\{\mu^2(\mu+1)^2-\nu^2(\mu-1)(1+3\mu)\}}{8(\nu^2+\mu^2)^3}.$$

3. *Discussion of results*

Functions (12), (13), (14) and (15) become simplified for exceptional cases. Taking account of Eq. (10) and (11) we see that in the case of sufficiently thick films with very large grain size $\mu \gg 1$ and $\nu \gg 1$, whereas for very thin, fine-grained films $\mu \ll 1$ and $\nu \ll 1$.

Considering $\Delta > 0$, i.e. $1/\mu^2+1/\nu^2 > 1$, we see that if $\mu > 1$, then $\nu < 1$. Hence, from Eq. (12) it follows that

$$\frac{\sigma_F}{\sigma_B} = \lim_{\mu\to\infty} W_1(\mu, \nu) = F_1(\nu),$$

where the function $F_1(\nu)$ is derived in Refs [16] and [17]. Function $F_1(\nu)$ holds for very thick films and small grain size ($\nu < 1$).

If $\Delta < 0$, i.e., $1/\mu^2+1/\nu^2 < 1$, then for $\mu > 1$ also $\nu > 1$. Using Eq. (15) we get

$$\frac{\sigma_F}{\sigma_B} = \lim_{\mu\to\infty} W_4(\mu, \nu) = F_2(\nu),$$

where the function $F_2(\nu)$ is also derived in Refs [16] and [17]. Function $F_2(\nu)$ holds in the case of very thick films and large grain size ($\nu > 1$). The functions $F_1(\nu)$ and $F_2(\nu)$ are similar to the Mayadas-Shatzkes grain boundary function [16, 17].

Considering a film without grain structure ($\nu \to \infty$), and using Eqs (12) and (15), we get

$$\frac{\sigma_F}{\sigma_B} = \lim_{\nu\to\infty} W_1(\mu, \nu) = C(\mu); \quad \frac{\sigma_F}{\sigma_B} = \lim_{\nu\to\infty} W_4(\mu, \nu) = C(\mu),$$

where $C(\mu)$ is Cottey's function [3] and describes the same effect as the Fuchs-Sondheimer function [1, 2].

It follows that for v tending to infinity $\lim_{v\to\infty} W_1(\mu, v) = \lim_{v\to\infty} W_4(\mu, v)$.

Function (14) is one of the solutions of the integral (9) for $\Delta = 0$; hence $1/\mu^2 + 1/v^2 = 1$. If $\mu \to \infty$ then $v = 1$; or if $v \to \infty$ then $\mu = 1$. Therefore, using Eq. (14), we obtain $\lim_{\mu\to\infty} W_3(\mu, v) = \frac{3}{8}(3\pi - 8) = 0.5343$. Thus $\varrho_F = 1.872\, \varrho_B$.

From Eq. (14) also follows that $\lim_{v\to\infty} W_3(\mu, v) = 3/4$; then $\varrho_F = 4/3\varrho_B$. The same magnitude is obtained from Eq. (13): $\lim_{v\to\infty} W_2(\mu = 1, v) = 3/4$ or from the function $C(\mu)$ after introducing $\mu = 1$.

The considerations presented lead to the following conclusions:

$$\lim_{\substack{\mu\to\infty \\ v\to 0}} W_1(\mu, v) = \lim_{v\to 0} F_1(v) = 0; \quad \frac{\sigma_F}{\sigma_B} = 0; \quad \varrho_F \to \infty,$$

$$\lim_{\substack{\mu\to\infty \\ v\to \infty}} W_4(\mu, v) = \lim_{v\to \infty} F_2(v) = 1; \quad \frac{\sigma_F}{\sigma_B} = 1; \quad \varrho_F = \varrho_B,$$

$$\lim_{\substack{v\to\infty \\ \mu\to 0}} W_1(\mu, v) = \lim_{\mu\to 0} C(\mu) = 0; \quad \frac{\sigma_F}{\sigma_B} = 0; \quad \varrho_F \to \infty,$$

$$\lim_{\substack{v\to\infty \\ \mu\to \infty}} W_4(\mu, v) = \lim_{\mu\to \infty} C(\mu) = 1; \quad \frac{\sigma_F}{\sigma_B} = 1; \quad \varrho_F = \varrho_B.$$

Fig. 4 shows ϱ_F/ϱ_B as a function of v and μ in terms of $W_n(\mu, v)$ given by Eqs (12), (13), (14) and (15), where $n = 1, 2, 3$ or 4. These functions combine both the internal

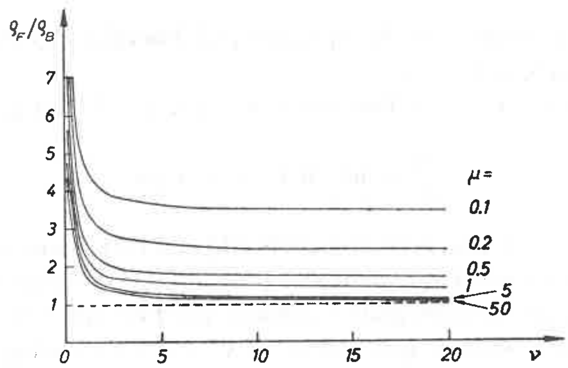

Fig. 4. ϱ_F/ϱ_B versus v and μ. The curves were plotted using Eqs (12), (13), (14) and (15)

and the external size effects considered. From Fig. 4 it follows that if $v \to 0$ or $\mu \to 0$, then $\varrho_F \to \infty$ and if $v \to \infty$ or $\mu \to \infty$ then $\varrho_F \to \varrho_B$.

In Fig. 5, ϱ_F/ϱ_B is plotted against grain diameter (in terms of λ_B) for given film thicknesses as well as for $r = 0.5$ and $p = 0.5$. The curves were developed by means of the calculation results obtained involving the function $W_n(\mu, \nu)$ together with Eqs (10) and (11).

As can be seen from Fig. 5, for small graon diameter or very thin film thickness, ϱ_F is considerably greater than ϱ_B; while for thicker films and large grain diameter, ϱ_F does not

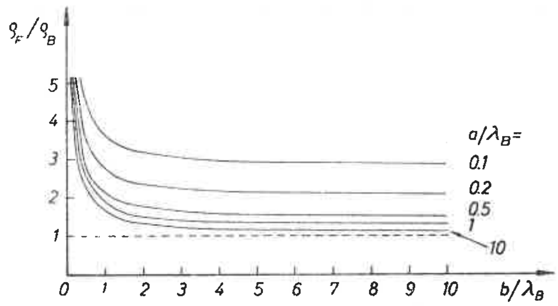

Fig. 5. ϱ_F/ϱ_B versus grain diameter and film thickness for $r = 0.5$ and $p = 0.5$. The curves were plotted using Eqs (12), (13), (14) or (15) together with Eqs (10) and (11)

significantly differ from ϱ_B. It may be easily shown that when p or r increase, ϱ_F decreases toward ϱ_B, because with increasing p or r, the values of μ or ν also increase (see Fig. 4).

The Mayadas-Shatzkes functions $\Phi(k, p, \alpha)$ [8, 14] is known to involve also external surface scattering and grain-boundary scattering effects. Mayadas and Shatzkes have taken into account Fuchs's size effect theory [1, 2] considering that the grains are bounded by the δ-function potentials. It is easy to notice that the values of ϱ_F/ϱ_B obtained using $W_n(\mu, \nu)$ or $\Phi(k, p, \alpha)$ for given film thicknesses and grain diameters are very close to each other. E.g., for $k \geqslant 1$, $\alpha < 0.5$ inconsistency is lower than 2%.

The dependence of resistivity on grain diameter has been proved experimentally for Al films [10] of a thickness of 600 Å and thicknesses ranging from 1800—3500 Å. The conformity of the theoretical curves is best at $r = 0.2$, $p = 0.1$ and $r = 0.5$, $p = 0.1$ for 600 Å thick and 2000 Å thick films, respectively.

Thanks are due to Professor C. Wesołowska for supervision, to Dr. S. Lewanowicz for computer calculations, and to Dr. E. Dobierzewska-Mozrzymas for helpful discussions.

REFERENCES

[1] K. Fuchs, *Proc. Camb. Philos. Soc.* **34**, 100 (1938).
[2] E. H. Sondheimer, *Adv. Phys.* **1**, 1 (1952).
[3] A. A. Cottey, *Thin Solid Films* **1**, 297 (1968).
[4] L. A. Moraga, A. Vilche, *Thin Solid Films* **38**, 117 (1976).
[5] S. Arajs, B. F. Oliver, J. T. Michalak, *J. Appl. Phys.* **38**, 1676 (1967).
[6] P. V. Andrews, M. B. West, C. R. Robeson, *Phil. Mag.* **19**, 887 (1969).
[7] V. P. Nagpal, V. P. Duggal, *Thin Solid Films* **9**, 313 (1972).
[8] A. F. Mayadas, M. Shatzkes, *Phys. Rev.* **B1**, 1382 (1970).
[9] P. Wissmann, *Thin Solid Films* **5**, 329 (1970).
[10] E. Dobierzewska-Mozrzymas, F. Warkusz, *Thin Solid Films* **43**, 267 (1977).

[11] G. Wedler, P. Wissmann, *Ber. Bunsenges. Phys. Chem.* **74**, 934 (1970).
[12] F. Thieme, W. Kirstein, *Thin Solid Films* **30**, 371 (1975).
[13] E. E. Mola, J. M. Heras, *Thin Solid Films* **18**, 137 (1973).
[14] E. E. Mola, J. M. Heras, *Electrocomp. Science and Technol.* **1**, 77 (1974).
[15] C. R. Tellier, A. J. Tosser, *Thin Solid Films* **33**, L19 (1976).
[16] F. Warkusz, *Electrocomp. Science and Technol.* (1978) (in press).
[17] F. Warkusz, *Kom. Inst. Fiz. Politechniki Wroclawskiej* 422 (1977), in Polish.