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Size effects of polycrystalline tin films

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The electrical properties of polycrystalline tin films evaporated onto a cooled quartz substrate were examined *in situ* and in vacuum. The relationship between internal size effect and annealing temperature was considered. It was found that the parameter α^* of grain-boundary scattering decreased as the annealing temperature was increased.

1. Introduction

Generally, thin metal films have higher resistivities than the bulk values. This is the result of external size effect and internal size effect, as well as the result of lattice imperfections which are more pronounced in thin films. The external size effect is defined as the effect of film thickness and electron scattering at the external surfaces on the electrical properties of the film, whereas the internal size effect is defined as the effect of grain diameter and grain-boundary scattering on the electrical properties of the film. The external size effect is described by the Fuchs–Sondheimer function $F(K, p)$ [1] and by Cottey's function $F(\mu)$[2], while the internal size effect is given by the grain-boundary scattering functions $G(\alpha)$ [3] and $F(\nu)$ [4].

In recent years some experiments on external and internal size effects have been reported [3, 5–13]. In this paper the influence of both internal and external size effects on the resistivity and temperature coefficient of resistivity (TCR) in tin films is considered in terms of the Mayadas–Shatzkes function [3] and of the function developed by Warkusz [4, 14].

2. Theory

2.1. Resistivity of thin films

Film resistivity can be described by the Mayadas–Shatzkes function [3]

$$\rho_f = \frac{\rho_0}{\Phi(K, p, \alpha^*)} \quad (1)$$

where ρ_0 is the resistivity of the bulk material $\Phi(K, p, \alpha^*)$ represents the Mayadas–Shatzkes function, $K = t/\lambda_0$, t is the film thickness, λ_0 is the electron mean free path in the bulk, p is the fraction of electrons specularly scattered at the external surfaces, $\alpha^* = \lambda_0 R/[D(1-R)]$ describes the parameter of grain-boundary scattering, D is the grain diameter, and R is the grain-boundary reflection coefficient.

If $K > 1$ and $\alpha^* < 1$, then $\Phi(K, p, \alpha^*)$ may be replaced by the product of Fuchs's function and grain-boundary scattering function $G(\alpha^*)$ [8, 15]

$$\Phi(K, p, \alpha^*) = F(K, p) \cdot G(\alpha^*). \quad (2)$$

Hence, Equation 1 can be written in a simplified form [8, 9, 15]:

$$\rho_f = \rho_0 \left\{ 1 + \frac{3}{8} \frac{\lambda_0}{t} (1-p) + \frac{3}{2} \alpha^* \right\}. \quad (3)$$

In the present paper, Equation 3 is a base for the interpretation of the experimental results.

The film resistivity can also be defined in the following way [14]

$$\rho_f = \frac{\rho_0}{W(\mu, \nu)} \quad (4)$$

where $W(\mu, \nu)$ refers to the function derived in [4] and [14]; $\mu = t/[\lambda_0 \ln(1/p')]$; $\nu = D/[\lambda_0 \ln(1/r)]$, and r is the coefficient of electron transmission through the grain boundary.

If $r = 1$, the electrons move through the grain boundary. If $r = 0$, the electrons are scattered at

the grain boundary. The relationship between the coefficients R and r may be expressed as $R/(1-R) = 0.62 \ln(1/r)$ or $\alpha^* = 0.62/\nu$ (cf. [4] and [14]).

If $\mu > 1$ and $\nu > 1$, then $W(\mu, \nu)$ can be substituted by the product of Cottey's function $F(\mu)$ and the grain-boundary scattering function $F(\nu)$ [4]

$$W(\mu, \nu) = F(\mu)F(\nu). \quad (5)$$

For $\mu > 1$ and $\nu > 1$, Equation 4 can take a simplified form:

$$\rho_f = \rho_0 \left\{ 1 + \frac{3}{8} \frac{\lambda_0}{t} \ln(1/p') + \frac{9}{32} \frac{\pi}{\nu} \right\}. \quad (6)$$

Because $\ln(1/p') = 1 - p' + 1/2(p'-1)^2 \ldots$, it follows that neglecting high-power terms, Equation 6 becomes identical with Equation 3, when $\nu = 0.62/\alpha^*$.

The second and third terms of the right-hand side of Equations 3 and 6 are responsible for the external and internal size effects, respectively [8, 15].

2.2. Temperature coefficient of resistivity (TCR) of thin films

A general expression for the TCR has the following form [16]:

$$\alpha_f = \alpha_0 - (\alpha_0 + \beta_0) \times \left\{ \frac{K}{F(K,p)} \frac{dF(K,p)}{dK} - \frac{\alpha^*}{G(\alpha^*)} \frac{dG(\alpha^*)}{d\alpha^*} \right\}, \quad (7)$$

which is true for $K > 1$ and $\alpha^* < 1$. In this equation, α_0 is the TCR of the bulk, β_0 the thermal expansion coefficient, and $\beta_0 \ll \alpha_0$ [17].

Using Equations 4 and 5 gives

$$\alpha_f = \alpha_0 - (\alpha_0 + \beta_0) \times \left\{ \frac{\mu}{F(\mu)} \frac{dF(\mu)}{d\mu} + \frac{\nu}{F(\nu)} \frac{dF(\nu)}{d\nu} \right\} \quad (8)$$

for $\mu > 1$ and $\nu > 1$.

Both Equation 7 and Equation 8 are derived by taking into account the external and internal (grain boundary) size effects.

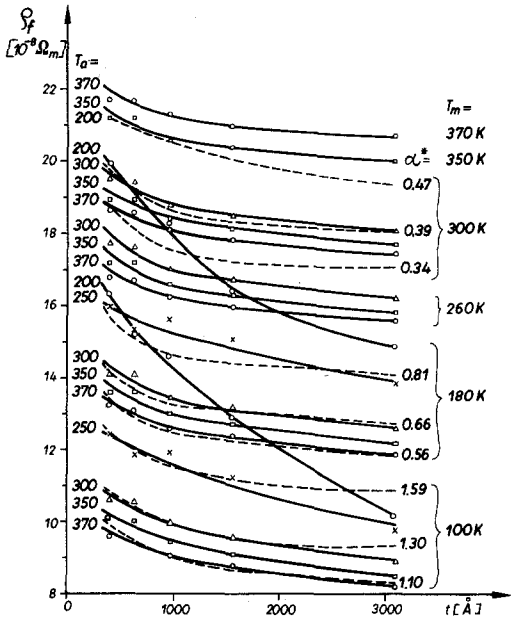

Figure 1 ρ_f versus t curves for $T_a = 200$, 250, 300, 350 and 370 K and $T_m = 100$, 180, 260, 300, 350 and 370 K.

3. Experimental details

Pure tin (99.999%) films were evaporated onto cooled fused quartz substrates in vacuum at $p_v \approx 10^{-6}$ Torr. The substrate temperature T_e was 142 K and the deposited films were 10 mm long and 0.5 mm wide. The evaporation rates were 14.1, 7.0, 4.3, 2.9 and 1.8 Å sec^{-1} and yielded film thicknesses of 3100, 1550, 950, 640 and 390 Å, respectively, which were measured in terms of Tolansky's method [18]. Film resistivities as a function of film thickness were determined using the method reported in [19]. The films were annealed at a temperature T_a of 200, 250, 300, 350 and 370 K for 0.5 h. The measurements of resistivity were performed at a temperature T_m between 100 and 370 K.

An electron transmission micrograph (Fig. 2) shows that the tin films obtained exhibit a polycrystalline structure (Fig. 3).

4. Discussion

Actual resistivities are represented in Fig. 1. Equations 3 and 6 are employed to fit the theoretical curves with the experimental results. Calculation of the theoretical curves is based on the values which are listed in Table I [20–23]. The fit of the theoretical (Fig. 1, dotted line) and experimental (Fig. 1, full line) curves shows

TABLE I The values of α^* and ν for tin films

T_m (K)	λ_0 (Å)	$\rho_0 \times 10^{-8}$ (Ωm)	T_a					
			250 K		300 K		370 K	
			α^*	ν	α^*	ν	α^*	ν
100	700	3.2	1.59	0.39	1.30	0.48	1.10	0.56
180	350	6.3	0.81	0.76	0.66	0.94	0.56	1.10
300	200	11.2	0.47	1.32	0.39	1.59	0.34	1.82

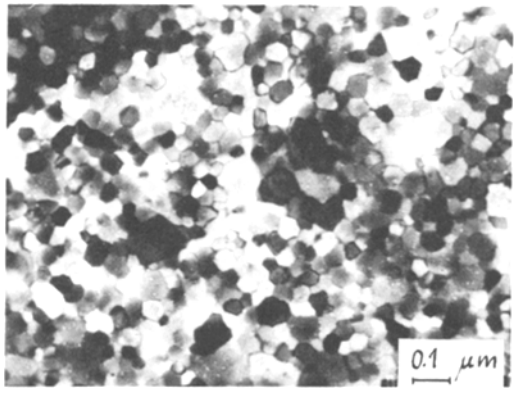

Figure 2 Transmission electron micrograph of 640 Å tin film.

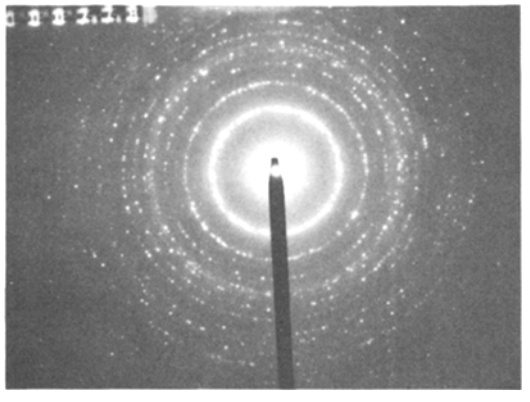

Figure 3 Electron diffraction pattern of 640 Å polycrystalline tin film.

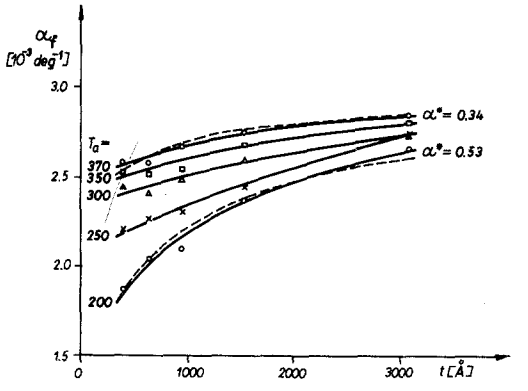

Figure 4 α_f versus t curves for a tin film with $T_a = 200$, 250, 300, 350 and 370 K.

that p is equal to 0.1 and α^* decreases with measuring temperature T_m; α^* is also decreased as the annealing temperature, T_a, increases. Some inconsistency between theoretical and experimental results was observed at low annealing temperatures. This seems to be attributable to a less-ordered grain structure.

The values of α^* and ν as a function of T_m and T_a, respectively, are also listed in Table I. The low α^* values at high annealing temperatures are an indication that the grain boundaries disappear [4, 8, 24].

For a 640 Å thick film the grain diameter D (Fig. 2, $D = 400$ Å) was measured, and the parameters R and r calculated. The results are as follows: at $T_a = 370$ K, $R = 0.39$ ($r = 0.36$); at $T_a = 300$ K, $R = 0.43$ ($r = 0.30$), and at $T_a = 250$ K, $R = 0.48$ ($r = 0.23$). The values of R are close to the ones reported for tin, aluminium, cobalt and nickel [6, 8, 9, 13].

In Fig. 4, the TCR of a tin film is plotted versus thickness and annealing temperature. The theoretical curves (dotted line) are fitted with the use of Equation 7.

A dependence of α_f on t similar to that plotted in Fig. 4 was obtained experimentally for tin [13] and other metal films [16]. Thus α_f was found to depend on the annealing temperature: with increasing T_a, the value of α_f is also increased. For thick tin films this dependence is linear.

5. Conclusions

(1) Film resistivity and TCR are functions of film thickness and annealing temperature; with increasing film thickness, annealing temperature and

annealing rate, film resistivity decreases [25] and TCR is increased.

(2) ρ_f and α_f are influenced by the grain structure. When T_a increases, α^* is decreased. This can be explained by the partial disappearance of the grain boundaries and the ordering of the grain structure which takes place at increased annealing temperatures.

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