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The study of temperature coefficient of resistivity of polycrystalline metal films

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The temperature coefficient of resistivity (t.c.r.) has been studied for the polycrystalline metal tin and lead films of various thicknesses. The t.c.r. is found to increase with thickness and thus exhibits the size effect. This thickness dependence of t.c.r. is successfully explained with the help of a three-dimensional model. The grain boundary t.c.r. and specularity parameter are determined from the measurements.

1. Introduction

The theory of electric conduction in thin metal films was developed by Fuchs [1] and Sondheimer [2]. In this theory the scattering of electrons within a film is described by a relaxation time, τ, and that at the surface of the film by a specularity parameter p, so as to permit an explicit solution of the Boltzmann transport equation.

Polycrystalline films have an additional contribution to the resistivity due to scattering of conduction electrons from grain boundaries [3]. Mayadas and Shatzkes [4] (MS) presented a model in which the total resistivity of a thin film was calculated from three types of electron scattering mechanisms: (1) an isotropic background scattering due to combined effect of phonons and point defects; (2) scattering due to external surfaces; and (3) scattering due to a distribution of planar potentials or grain boundaries.

The resistivity of polyvalent metals such as tin and lead films have been the subject of much research in the past. The electrical properties like resistivity, temperature coefficient of resistivity (t.c.r.) of metallic tin and lead films have been studied by several authors [5–10]. Chandra and Katyal [11, 12] have also studied the electrical resistivity and t.c.r. of thin metallic tin and lead films of various thicknesses in a temperature range of 150 to 300 K. They observed that resistivity is consistent with the Mayadas model for completely diffuse scattering, i.e. $p = 0$.

Recently [13] we have calculated the electrical resistivity due to the scattering of conduction electrons from the grain boundaries by considering completely diffuse and partially diffuse scattering in polycrystalline films of lead, tin and tin–lead alloys. By using a theoretical equation derived from MS model it was shown that the grain boundary resistivity decreases with the increase in grain diameter and the effect of the specularity parameter, p, on it is small. In the present work the temperature coefficient of resistivity has been studied for polycrystalline tin and lead films of various thicknesses. The grain boundary t.c.r. and specularity parameter p have been determined from the experimental results.

2. Experimental procedure

The experimental procedure for the preparation and electrical conductivity measurements of tin and lead films have already been described [11, 13]. All the samples of tin and lead (99.999% pure) were thermally evaporated in a vacuum better than 10^{-5} torr. The glass substrate was kept at room temperature during the evaporation for all the samples. All the electrical measurements of the samples kept in a vacuum better than 10^{-5} torr were made with the help of K-3 potentiometer. The four silver electrodes (silver 99.99% pure) were used as contacts. During the electrical measurements, the temperature of the substrate was varied by a cold finger filled with liquid nitrogen and fitted with a heater at the bottom near the substrate. The thickness and evaporation rate were measured by a calibrated quartz crystal thickness monitor (Model CFM-1 Hind Hi Vac). The grain size of tin films was studied by a scanning electron microscope while the grain size of lead films was determined by X-ray studies.

3. Experimental results and discussion

The study of tin [11] and lead [12] films under the scanning electron microscope and X-ray technique shows that films are polycrystalline in nature. The temperature coefficient of resistivity of tin and lead films increases with film thickness and thus exhibits the size effect (Figs 1 and 2).

To consider the effect of grain boundary scattering on the transport properties of metal films. Mayadas and Shatzkes [4] assumed that grain boundaries could be represented by geometrical arrays of planes parallel as well as perpendicular to the applied electric field. On the basis of models established earlier [14] to describe electron scattering by dislocation, these authors have assumed that the scatterers parallel to the electric field have no influence on the conductivity. Using this oversimplification they have considered only the effect due to planes perpendicular to the electric field. Thus, this procedure adopted by Mayadas and Shatzkes to calculate film conductivity gives a one-dimensional model which is not suitable for

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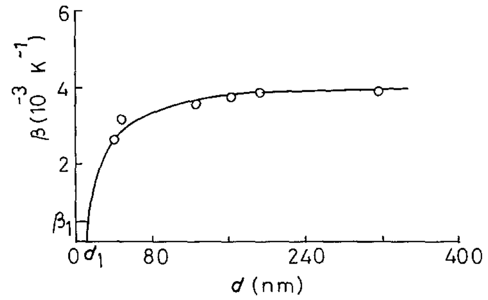

Figure 1 Plot of t.c.r. against thickness for tin films.

analysing the transport phenomena occurring in three-dimensional space. Pichard *et al.* [15] proposed a three-dimensional model to express the resistivity of metal films. In this model it is assumed that grain boundaries in polycrystalline films can be represented by three arrays of mutually perpendicular planar potentials. The average effect of grain boundary is represented by a specular transmission coefficient, t, 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 [15, 16]. Assuming that the probability of an electron travelling a given distance, without being diffusely scattered, is given by an exponential law [16], a mean free path can be ascribed to the three-dimensional array of scatterers whose spacing is identified with the average grain diameter, D. They further introduced a parameter, ν, known as grain boundary parameter, defined as [15]

$$\nu = \frac{D}{l_0}\left[\ln\left(\frac{1}{t}\right)\right]^{-1} \tag{1}$$

where l_0 is the bulk mean free path.

The ratio of infinitely thick polycrystalline film conductivity to that in bulk material was found to be [15]

$$\frac{\sigma_g}{\sigma_0} = \frac{\varrho_0}{\varrho_g} = \frac{3}{2}\frac{\nu}{1-C}\left[r - \tfrac{1}{2} + (1 - r^2)\ln(1 + r^{-1})\right] \tag{2}$$

with

$$r = \frac{\nu + C^2}{1 - C}; \quad C = \frac{4}{\pi}$$

where ϱ_0 is the bulk resistivity and ϱ_g the resistivity of infinitely thick polycrystalline film where both background scattering and grain boundary scattering are operative.

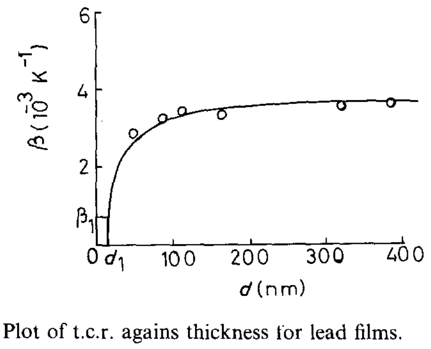

Figure 2 Plot of t.c.r. agains thickness for lead films.

With the consideration of partially diffuse scattering, the external size effect can be analysed in terms of Cottey's model [17] in which a mean free path is associated with scattering; an "external surface" parameter, μ, can be introduced to describe the size effect, i.e.

$$\mu = \frac{d}{l_0}\left(\ln\frac{1}{p}\right)^{-1} \tag{3}$$

where d is the film thickness. Consequently, in thin polycrystalline films in which three types of electron scatterings, i.e. background scattering, grain boundary scattering and external surface scattering, are simultaneously operative, the electrical conductivity σ_{Fp} takes the simple analytical form [18]

$$\frac{\sigma_{Fp}}{\sigma_0} = \frac{3}{2}\frac{1}{b}\left[\alpha - \tfrac{1}{2} + (1 - \alpha^2)\ln(1 + \alpha^{-1})\right] \tag{4}$$

with

$$b = \mu^{-1} + \nu^{-1}(1 - C) \tag{5}$$

$$\alpha = (1 + C^2\nu^{-1})b^{-1} \tag{6}$$

This three-dimensional model is very similar to the MS model [4]. For any type of film structure (polycrystalline or columnar or monocrystalline) Equation 4 holds and can be regarded as an alternative algebraic formulation for the complicated expression obtained by Mayadas and Shatzkes [19].

To analyse the data on film t.c.r. in terms of size effect theories the following assumptions are made

(i) the rigid band model of metals is valid,

(ii) the number of conduction electrons per unit volume is temperature independent,

(iii) the thermal expansions of the grains and film's dimensions are negligible in comparison to that of mean free path.

In the framework of the three-dimensional model, Pichard *et al.* [20, 21] studied the t.c.r. of polycrystalline film. From Equation 2 they derived an equation for grain boundary t.c.r., β_g, as

$$\frac{\beta_g}{\beta_0} = \frac{\nu}{1 - C}\,\frac{r^{-1} - 2 + 2r\ln(1 + r^{-1})}{r - \tfrac{1}{2} + (1 - r^2)\ln(1 + r^{-1})} \tag{7}$$

In the case when $\nu \gg 1$ this equation becomes

$$\frac{\beta_g}{\beta_0} \approx 1 + \frac{1}{\nu}\left[\tfrac{3}{8}(1 - C) - C^2\right] \tag{8}$$

while the total film t.c.r. β_{Fp} was expressed as [20]

$$\frac{\beta_{Fp}}{\beta_0} = \frac{1}{b}\,\frac{\alpha^{-1} - 2 + 2\alpha\ln(1 + \alpha^{-1})}{\alpha - \tfrac{1}{2} + (1 - \alpha^2)\ln(1 + \alpha^{-1})} \tag{9}$$

where β_0 is the bulk t.c.r.

To explain our experimental results on t.c.r. with the help of this model, we have used the idea of two layer structure proposed by Tellier and Tosser [22]. The thickness dependence of the electrical resistivity of annealed and non-annealed films has provided evidence of the existence of two layers within the metallic films [22]. This is in agreement with earlier described nucleation growth model; the bottom layer

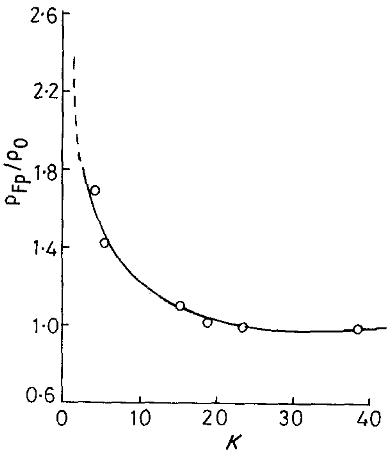

Figure 3 Variation of $\varrho_{\mathrm{Fp}}/\varrho_0$ with reduced thickness, K, for tin films.

has a constant thickness d_1 known as the first critical thickness and electric conduction according to Mayadas and Shatzkes [4] occurs in the upper layer of thickness d_2. Since the total conductivity is the sum of reciprocal resistance r_1' and r_2', the t.c.r. of the second layer β_2 is calculated from the relation [23]

$$\beta_2 = \beta + (\beta - \beta_1)(r_2'/r_1') \qquad (10)$$

where β is the total film t.c.r.; β_1 and r_1' are the t.c.r. and resistance respectively of d_1; r_2' is the resistance of d_2.

The critical thickness has been calculated from the experimental data on resistivity of tin [11] and lead [12] as $d_1 = 12\,\mathrm{nm}$ for tin and $d_1 = 17\,\mathrm{nm}$ for lead. Figs 1 and 2 give the value of t.c.r of d_1 as $\beta_1 \approx 0.5 \times 10^{-3}\,\mathrm{K}^{-1}$ for tin and $\beta_1 \approx 0.7 \times 10^{-3}\,\mathrm{K}^{-1}$ for lead.

For the calculation of resistance r_1', the value of ϱ_1 has been deduced from the variation of $\varrho_{\mathrm{Fp}}/\varrho_0$ with the reduced thickness $K (K = d/l_0)$. At a reduced thickness $K_1 = d_1/l_0$, $\varrho_{\mathrm{Fp}}/\varrho_0$ will become ϱ_1/ϱ_0. From Fig. 3 and taking $\varrho_0 = 12.6\,\mu\Omega\,\mathrm{cm}$, $l_0 = 8.3\,\mathrm{nm}$ [11] and $d_1 = 12\,\mathrm{nm}$, the value of ϱ_1 is calculated as $25.7\,\mu\Omega\,\mathrm{cm}$ for tin films. While for lead films for which $\varrho_0 = 24.4\,\mu\Omega\,\mathrm{cm}$, $l_0 = 4.3\,\mathrm{nm}$ [12] and $d_1 = 17.0\,\mathrm{nm}$, Fig. 4 gives the value of ϱ_1 as $55.0\,\mu\Omega\,\mathrm{cm}$. If ϱ_1 is known then the resistance of the first layer r_1' can be calculated by the relation $r' = \varrho L/ld$, L and l are the length and width of the films respectively.

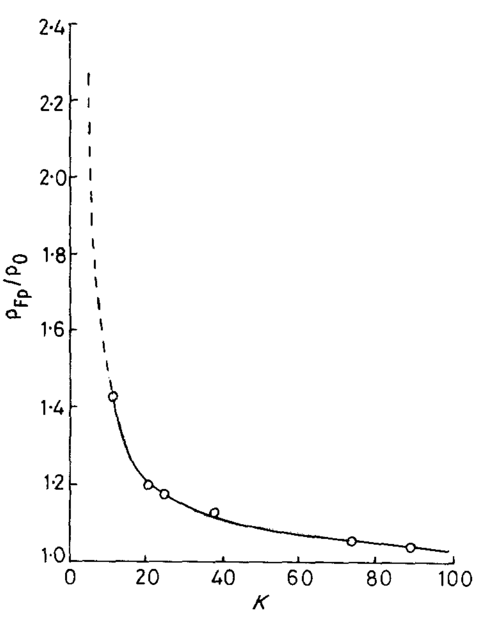

Figure 4 Variation of $\varrho_{\mathrm{Fp}}/\varrho_0$ with reduced thickness, K, for lead films.

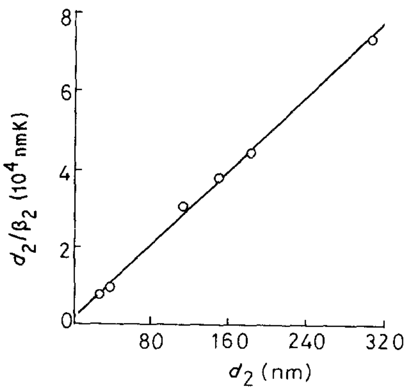

Figure 5 Plot of d_2/β_2 against thickness, d_2, of second layer for tin films.

Figs 5 and 6 show that the plot of d_2/β_2 against d_2 yields a straight line of slope β_g^{-1} as expected [20]. From Figs 5 and 6 we obtained $\beta_g \approx 4.07 \times 10^{-3}\,\mathrm{K}^{-1}$ for tin films; and $\beta_g \approx 3.87 \times 10^{-3}\,\mathrm{K}^{-1}$ for lead films.

We may determine from Equations 1 and 7 the experimental value of the grain boundary parameter v. Taking $\beta_0 = 4.35 \times 10^{-3}\,\mathrm{K}^{-1}$ for tin films [7], and $\beta_0 = 4.3 \times 10^{-3}\,\mathrm{K}^{-1}$ for lead films [24] we obtained $v = 24.0$ for tin films and $v = 15.0$ for lead films.

These experimental values of v and β_g and usual bulk mean free path value were used to determine the theoretical variations of β_2 with thickness d_2 for the different values of the specularity parameter p. The numerical values of $\beta_{\mathrm{Fp}}/\beta_0$ (Equation 9) have been calculated using a DEC 10 computer. Figs 7 and 8 show the theoretical thickness dependence of t.c.r. and experimental data.

From the analysis of these various curves the following conclusions can be made.

1. Figs 7 and 8 for tin and lead, respectively, show that there is a good agreement between the experimental data and theoretical variation from Equation 9. The best fit is found for $p = 0.2$ in the case of tin films and $p = 0.01$ for lead films. The low values of the specularity parameter p were expected because the films were not thoroughly annealed.

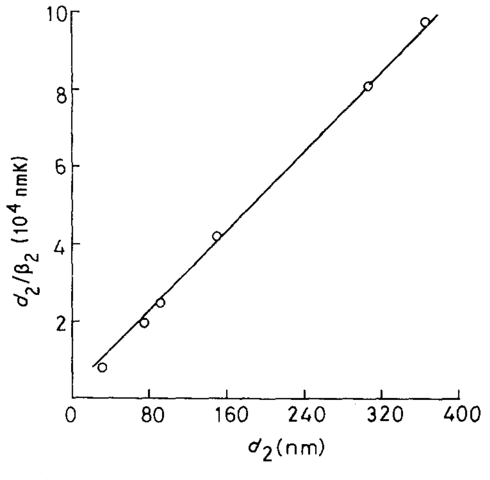

Figure 6 Plot of d_2/β_2 against thickness, d_2, of second layer for lead films.

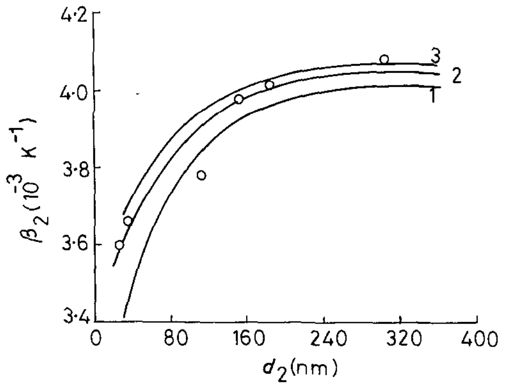

Figure 7 Theoretical thickness dependence of t.c.r. β_2 of tin films from Equation 9 for $\nu = 24.0$ and the experimental data (1) $p = 0.1$, (2) $p = 0.2$, (3) $p = 0.3$.

2. As no marked discrepancies are observed in the values of p, the thickness dependence of t.c.r. is well understood. Therefore, it can be assumed that two-layer model [22] and t.c.r. model [20] give a suitable description of the t.c.r. of polycrystalline metal films.

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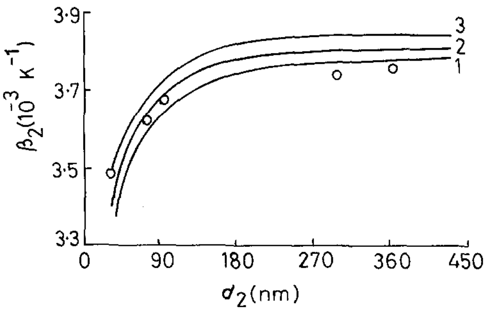

Figure 8 Theoretical thickness dependence of t.c.r. β_2 of lead films from Equation 9 for $\nu = 15.0$ and the experimental data (1) $p = 0.01$, (2) $p = 0.02$, (3) $p = 0.05$.

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