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Effect of grain-boundary scattering on the electrical resistivity of indium films

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Electrical resistivity and temperature coefficient of resistivity of polycrystalline indium films have been studied in the temperature range 30 to 90° C. It has been observed that the grain-boundary scattering theory of Mayadas and Shatzkes reproduces the experimental observations more faithfully than the size-effect theory given by Fuchs.

1. Introduction

Generally, polycrystalline films deposited onto glass substrates have much higher resistivities than the bulk values. Data analysis is usually made using Fuchs relations [1] by adjusting the specularity parameter, the bulk resistivity and the mean free path being assumed to be thickness independent. In practice, the structure of a thin film does not approximate to a single crystal plane parallel slab but rather consists of an array of randomly oriented polycrystallites. The crystal structure is often of a columnar type in which the individual crystals grow roughly vertically from the substrate to the upper surface of the film. Consequently, the only boundaries which need to be considered are those lying parallel or perpendicular to the applied field. The linear dimension of the crystallites in the plane of the film often becomes comparable with the electron mean free path and consequently an additional deviation from Matthiessen's rule due to scattering at the boundaries of the crystals should be taken into account. Mayadas and Shatzkes [2] evaluated the problem in a simplified way (henceforth M–S theory) to allow a qualitative description of grain-boundary scattering in polycrystalline films which more or less faithfully reproduced the experimental observations on aluminium [2], tungsten [3], cobalt [4] and bismuth [5] films.

Earlier works worth mentioning are due to Aleksandrov [6] and Olsen [7]. Aleksandrov studied the size dependence of residual resistance using the sample in wire form. Magneto-resistance and size effect in indium wires was studied by Olsen. Marked deviation from Matthiessen's rule was observed. Recently, Reale [8] determined the size effect parameters of indium from his measurements of Hall effect and sheet resistivity at helium temperature. In this paper we have reported *in situ* measurements of resistivity and temperature coefficient of resistivity (TCR) of indium films (500 to 6000 Å) at temperature between 30 and 90° C.

2. Experimental

Pure indium (99.999%), Johnson and Matthey, UK) was deposited from a molybdenum boat on to a cleaned glass substrate at room temperature. Prior to deposition the substrate was baked at 150° C for 2h at a pressure of $\sim 10^{-6}$ Torr. Potentiometric measurements were carried out *in situ* using four-probe method by van der Pauw [9] and the electrical leads were connected to the substrate by zinc–tin solder. The rate of deposition as monitored by a quartz crystal thickness monitor was always between 5 and 10 Å sec^{-1}. Actual thicknesses were measured by a multiple beam interferometer with an accuracy of 30 Å. The temperature was controlled by an electronic on–off temperature controller (± 1° C) and the film temperature was measured by a copper constantan thermocouple placed on the glass substrate. The maximum rise in temperature during deposition was always between 2 and 4° C.

3. Results and discussion

Resistivities of indium films (500 to 6000 Å) as a function of thickness at temperature (30, 60 and 90° C) are shown in Fig. 1.

It has been observed (Fig. 2) that the TCR of the film changes from a negative value to a positive one at about 700 Å before attaining the bulk value at higher thicknesses. This is probably due to the island structure of the films at initial stages of growth as revealed by the electron micrograph (Fig. 3). The negative TCR at low thicknesses could easily be understood from their island structure. As the film thickens the incoming atoms do not see the substrate as such and the islands become covered with polycrystalline over-layers. Therefore, with increased thickness the main bulk of the film would consist of smoother polycrystalline layers and thus the effect of under layers with island structure becomes less and less predominant. At some thickness the under-layer becomes very small in comparison with the total bulk of the film and the film then behaves bulk-like with positive TCR. At this point it is interesting to study the annealing effect of the films with

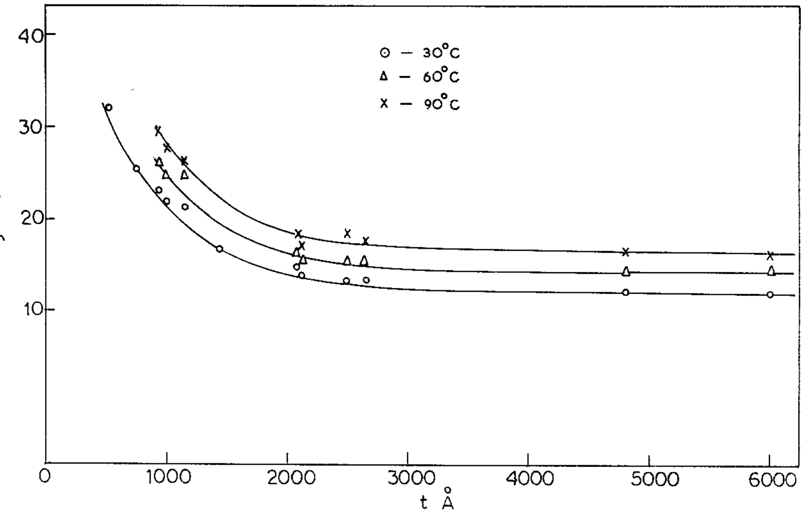

Figure 1 Resistivities of indium films as a function of thickness at temperatures of 30, 60 and 90° C.

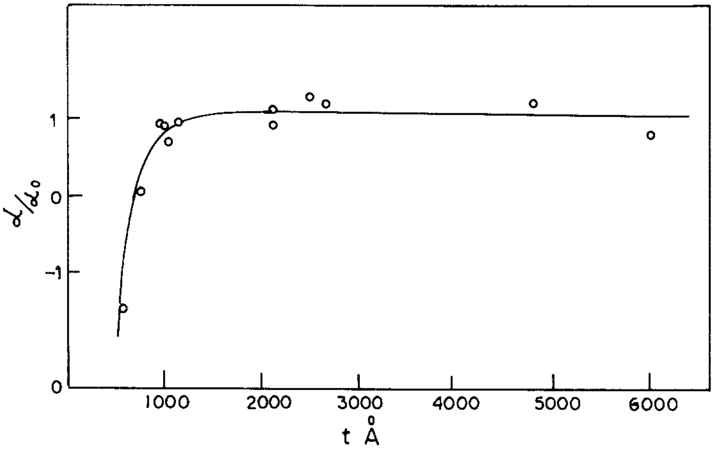

Figure 2 Plot of α/α_0 versus t.

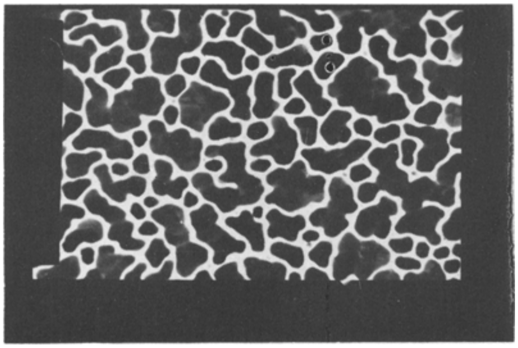

Figure 3 Transmission electron micrograph of indium film (~ 800 Å) × 12 000.

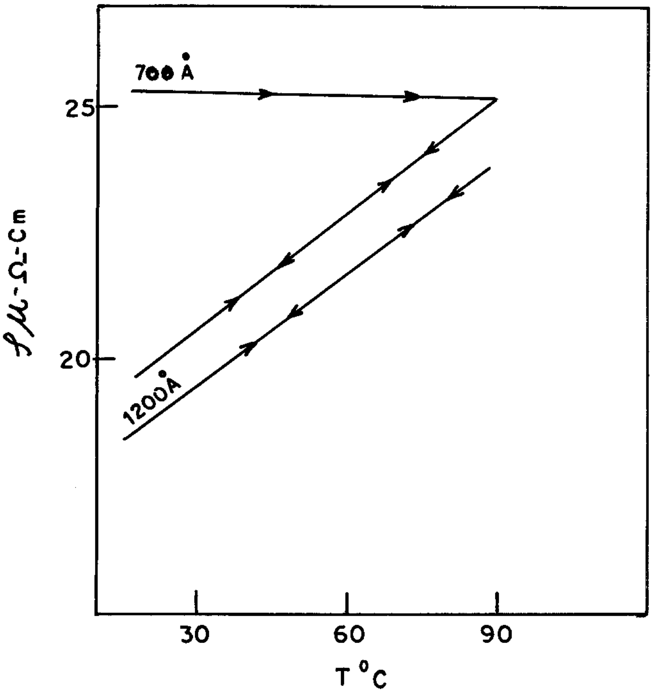

Figure 4 Effect of annealing on films (700 and 1200 Å) followed by subsequent thermal cycling.

subsequent thermal cycling. The effects of annealing (at 90°C for 2h) and subsequent thermal cycling on films of thickness 700 and 1200 Å are shown in Fig. 4. The thickness around 700 Å seems to be the critical thickness of indium films beyond which the films behave as a bulk. At about 700 Å, the islands, probably with a small over-layer, seem to coalesce with increasing mobility with temperature and the films show positive TCR. The 1200 Å thick film shows a predominant bulk-like behaviour due to a sizable amount of over-layer on the initial growth structure. With the coalescence of the islands, the physical structure of the film resembles a network and the electrical conduction is then determined by the islands, bridges and gaps between the islands. Electrical conductance is very sensitive to the changes brought about by ageing, annealing and adsorption. A great variety in the behaviour of the temperature dependence of resistance may, therefore, occur [10]. As the net work structure fills up, the porosity decreases and the conductivity of porous film is determined largely by grain-boundary scattering, diffuse scattering at grain surfaces and intergranular tunnelling.

Using the Boltzmann transport equation and assuming a spherical Fermi surface, an isotropic electron mean free path and partial specular scattering of the electrons from the surface of a film, Fuchs [1] obtained the ratio of resistivity of bulk to thin films as

$$\frac{\rho}{\rho_0} = \left[1 - 3(1-p)/2\gamma \int_1^\infty (a^{-3} - a^{-5})(1 - e^{-\gamma a}) /(1 - pe^{-\gamma a})\,da\right]^{-1} = \phi(p,\gamma) \qquad (1)$$

where ρ_0 is the resistivity of the bulk material, ρ is the resistivity of the thin film, γ is the ratio of the film thickness t to the mean free path l in the bulk, p being the specularity parameter.

A convenient form of this expression for the resistivity has been given by Chambers [11] and Sondheimer [12]. The limiting form of resistivity and TCR is given by

$$\frac{\rho}{\rho_0} = 1 + \frac{3}{8\gamma}(1-p) \qquad (2)$$

$$\frac{\alpha}{\alpha_0} = 1 - \frac{3}{8\gamma}(1-p) \qquad (3)$$

where α is the TCR of film and α_0 is the TCR of bulk material. Equations 2 and 3 indicate linear dependence of ρ and α on $1/t$ and the experimental plots of $\rho_{30^\circ\mathrm{C}}$ and $\alpha_{30^\circ\mathrm{C}}$ versus $1/t$ (Fig. 5) yield the values of $l(1-p)$ equal to 995 and 924 Å, which are in good agreement with each other. The values of ρ_0 and α_0 obtained from the intercepts are 11.87 $\mu\Omega$ cm and 0.00597° C^{-1} respectively. The higher value of bulk resistivity of thick films may be due to the fact that the indium films are

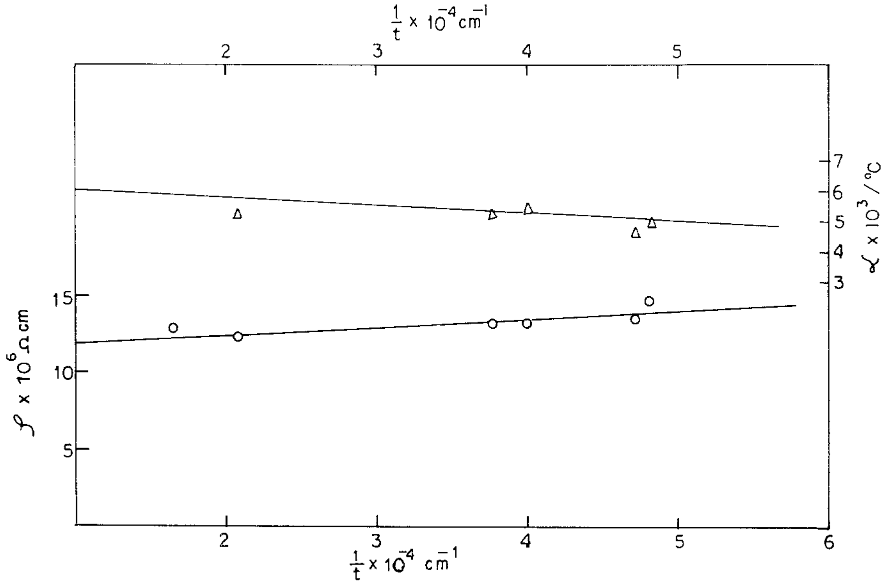

Figure 5 Dependence of resistivity and temperature coefficient of resistivity on reciprocal thickness. ⊙, Experimental resistivity data; △, experimental TCR data. Solid lines are the least squares fit straight lines.

not really planar, in fact the island structure shown in Fig. 3 ranges from 1500 to ~6000 Å.

Mayadas and Shatzkes [2] initiated the idea of grain-boundary scattering in polycrystalline films and evaluated the problem by considering the resistivity caused by the scattering of conduction electrons by grain boundaries together with the normal background scattering caused by defects and phonons. Mayadas and Shatzke's expression for film resistivity can be expressed as

$$\frac{\rho}{\rho_0} = \frac{\phi(p, \gamma_i)}{f(\alpha')}$$

where

$$f(\alpha') = 3\left[\frac{1}{3} - \frac{\alpha'}{2} + \alpha'^2 - \alpha'^3 \ln\left(1 + \frac{1}{\alpha'}\right)\right]$$

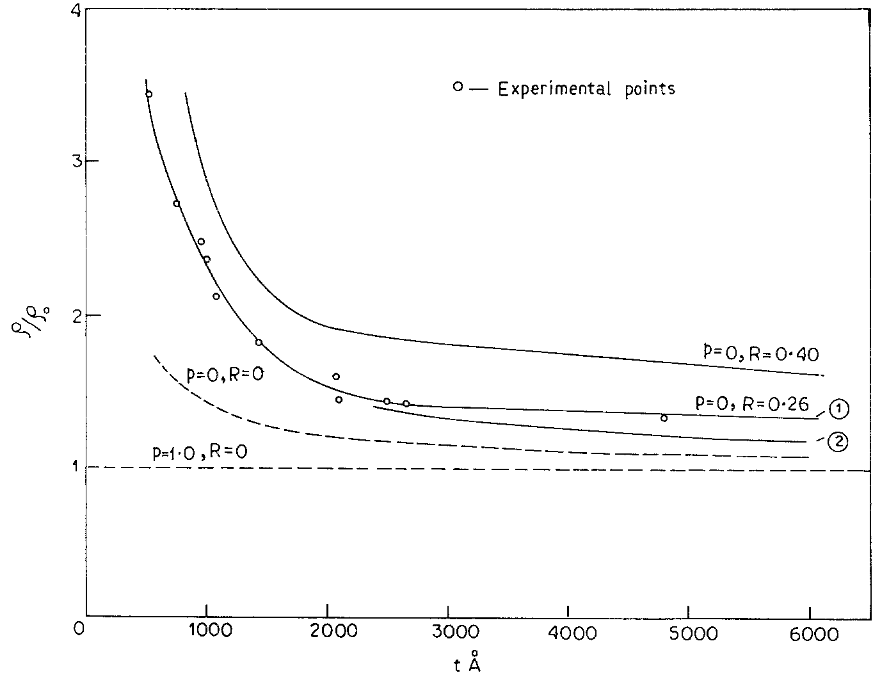

Figure 6 Plot of ρ/ρ_0 versus t. ⊙, Experimental point; ---- Fuchs curve ($R = 0, p = 0; R = 0, p = 1$); —— M–S curve, $R = 0.26$, $p = 0$ (curve 1: assuming grain size becoming constant beyond 1700 Å; curve 2: assuming grain size increases with thickness), and $R = 0.40$, $p = 0$.

α' being equal to $lR/D(1-R)$. $\phi(p,\gamma_i)$ is given by Equation 1 in which γ is replaced by $\gamma_i = \gamma/f(\alpha')$. D is the grain size and R is the coefficient of reflection of the grain boundary.

For comparison, the grain size is generally taken to be equal to the thickness of the film which should be true for small thicknesses. The grains do grow with thickness but not indefinitely such that the grain size becomes nearly constant as the film thickens. A curve corresponding to $p = 0$ and $R = 0.26$ using grain size equal to thickness is shown in Fig. 6 (curve 2). The agreement between the experimental curves and the M–S curve is quite good up to a certain range corresponding to a thickness of 1700 Å, after which the two curves substantially deviate from each other. At this point, it is very interesting to note that if we assume the grain size to be constant beyond 1700 Å, the M–S curve reproduces the experimental data faithfully over the entire range of measurement (curve 1, Fig. 6). Fuchs curves corresponding to two extreme cases ($p = 0$ and $p = 1$) are also shown in Fig. 6. The disagreement of our data with the size-effect theory for all thicknesses is quite apparent from Fig. 6.

It was also observed that the films (above 1000 Å) subjected to annealing at 90°C followed by subsequent thermal cycling show reversible resistivity behaviour with temperature. Also deposition at high substrate temperature (up to 90°C) has no effect on the resistivity of indium films.

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