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ON THE ELECTRICAL RESISTIVITY OF POLYCRYSTALLINE Al-FILMS

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Abstract:

Thickness dependence of the electrical resistivity of evaporated polycrystalline aluminium thin films have been studied. Films from 70 to 3000Å were deposited at room temperature on glass substrates. The electrical resistivity was measured in situ at room temperature by the Van der Pauw four-prope method. It was found that the experimental data fit faithfully the Mayadas-Shatzkes theory with the specular parameter P equals zero and the reflection coefficient at the grain boundary R equals 0.05 and 0.15 for deposition rates equal 2 and 7 Å/S, respectively. This indicates that the scattering mechanism due to the grain boundaries is affected by the deposition rate. It is weak at low deposition rates and becomes some what significant at higher rates. For oblique incident more than 45° the resistivity increases but Fuchs-theory is still applicable.

Introduction:

Electron transport phenomena in two dimentional solids has motivated extensive investigations since end of the nineteenth century. The size effects due to thickness (size) were subjected to intensive study theoretically and experimenally. The electric resistivity of a thin film is found to be many order of mangitude larger than that of the bulk material. Electric conductivity of a metal is directly proportional to the mean free path (mfp). As the thickness of a metal film becomes comparable in magnitude with mfp the film boundaries imposes a geometrical limitation on the movement of the conduction electrons.

Many size-effect theories(1,2), since Thomson in 1901, were proposed to explain the observed low electric conductivity of thin films as compared with that of the bulk. Among the various transport properties, such as the temperature coefficient of resistivity, Hall coefficient and thermoelectric power, the electrical resistivity(3) shows the best agreement with Fuchs and Sondheimer (F-S) theory. As extension to F-S theory, Mayadas and Shatzkes(2) have taken into account scattering of carriers by grain boundary surfaces especially at a grain size smaller than the mfp of conduction elections.

Somewhat contrary data on polycrystalline thin films have been given in literature. It was found by Mayadas et al.(4-6) that their data could not fit to the Fuchs curve with a single value of the specularity parameter P and bulk resistivity ρ_o but could be interporeted in terms of decrease in ρ_o with increasing thickness. Suri, Thakoor and Chopra(7) found, for Cu films deposited with a rate equals 6-8 Ås^{-1}, that the data depart markedly from the prediction of both F-S and the M-S theories. Mayadas and Shatzkes(2) and Bandyopadhyay and Pal(8) have studied Al-films and concluded that the grain boundary scattering is quite appreciated. However, they found different values for the reflection coefficient (R) at the grain boundaries, that is 0.17 and 0.28, respectively. Mayadas and Shatzkes did not specify the deposition rate while Bandyopadhyay and Pal have used rate equals 10 Å s^{-1}. So that this work aims to study the fitting of both F-S and M-S theories at different deposition rates and angles for Al-films. Also the effect of the deposition rate on the contribution from the grain boundaries scattering and the applicability of the theories at relatively slow rates are investigated.

Experimental:

Polycrystalline aluminium-films were prepared by thermal evaporation of high-purity Al (99.999%) from a tungesten helix. Films from 70 to 3000 Å were deposited onto glass substrates at room temperature in a vacuum better than 10^{-5} Torr. The substrates were washed ultrasonically and cleaned in the deposition champer by ionic bombardment. Deposition was carried out at different angless between the normal to the substrate and the deposition direction, namely zero, 45° and 60°. The rates of deposition used were 2 and 7 Å s^{-1} and they were carfully controlled. The rate and thickness were monitored by a quartz crystal oscillator. Calibration was carried out against interferometrically(9) known film thicknesses.

The electric resistivity was measured in situ at room temperature by the Van der Pauw(10) method. Thin copper vires were soldered to the glass substrate by spec-pure indium before deposition. Current-voltage characteristic was measured to examin the performance of the contacts.

Results and Discussion:

The performance of the indium contacts (electrodes) was examined by investigating the I-V characterestics as shown in Fig. (1). The experimentally obtained linearity assertained the needed ohmic contacts.

Data analysis in thin films is carried out assuming that the scattering mechanism is not bulk like and deviation from Mathiessen's rule is produced. According to Fuches size-effect theory, additional contribution to resistivity may rise because of surface scattering from the film surface (3). Based on the Fuch's approach, Chambers(11) and Sondheimer(1) gave the following convenient form of the expression for resistivity of thin film:

$$\frac{\rho}{\rho_o} = \phi(P, k) = 1 + \frac{3}{8k}(1 - P) \qquad (2)$$

where ρ_o is the thickness-independent bulk or intrinsic resistivity, k is the ratio of the film thickness, t, to the electron mfp, ℓ_o, in the bulk. P is the probability that an electron will be specularly reflected upon scattering from a film surface (specular parameter) and takes values from 0 to 1.

In order to apply F-S theory, values of ρ_o and (l-p) should be found. Plotting the linear relation between ρ t and t gave the straight line shown in Fig. (2). From the slop and intercept ρ_o and ℓ(l-p) were calculated, respectively. The experimental results yield the value of ρ_o equals 5.21 x 10^{-8} Ωm and ℓ_o(l-p) equals 421 Å for deposition rate of 2 Å/S. Fig. (3) shows the nomalized electric resistivity, ρ/ρ_o, versus film thickness. The theortical curve (continuous line) was drawn using eqn.(2) and the above estimated values. Fitting of the experimental data to the F-S theory is found to be reasonably accepted. In literature they assumed that eqn.(2) can be reduced to a family of ρ versus t curves with P as an adjustable parameter. This is meaningless because the values which are obtained for the best fit are estimated from the constants of the straight line shown in Fig.(2). Such straight line yields the product ℓ_o(l-p) and by this method neither the value of the mean free path, ℓ_o, nor the value of the specular parameter, P, can be obtained independently. Any change in P leads to a change in ℓ_o while the product ℓ_o(l-p) remains constant. Therefore no change in the shape of the theoritical curve is expected.

It was found this work and in literature(8) that both ρ_o and ℓ_o were more large, nearly douple, than that of the bulk material. The large value of is expected due to the large amount of lattice defects in Al-films as compared with the strain free bulk materials. If the specularity factor is taken larger than zero, the value of mfp will increase which is unreasonable. Therefore, the mfp, ℓ_o, is not derivable from the resistivity, ρ_o, although they are not independent. They related to each other by the following relation.

$$\rho_o \ell_o = (12\pi^3 \hbar) / (e^2 S)$$

where S is the Fermi surface area and e is the electronic charge. Mayadays et al.(4) suggested that the product $\rho_o \ell_o$ is a material constant and essentially independent of temperature. Others(8) expected higher $\rho_o \ell_o$ values at higher temperatures. Different values of $\rho_o \ell_o$ were given in literature, namely 31.67 x 10^{-12} Ωcm², and 8.2 x 10^{-12} Ωcm² for thin films at 323 and 4.2 K, respectively. For the bulk at 300 K, the value was 10.07 x 10^{-12} Ω cm². The results of the present work gave 21.9 x 10^{-12} Ωcm² at room temperature ($\cong$ 300 K). This means that the Fermi surface area decreases with temperature and at the same temperature its values for thin film is lower than that for the bulk.

In addition to the normal scattering of conduction electrons by phonons and lattice defects, and at the film surface, Mayadas and Shatzkes(5) considered the scattering at the grain boundaries. So the grain boundary contribution should produce additional deviation from Mathiessen's rule. The grain boundary scattering modifies expression(2) as folows:

$$\frac{\rho}{\rho_o} = \frac{\Phi(P,\ k')}{f\ (\alpha)}\ \left[\frac{1}{f(\alpha)} + \frac{3}{8k}\ (1-p)\right]$$

where $k' = k/f(\alpha)$,

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

ρ_g = the conductivity in the presence of both grain boundery and back ground scattering, and

$$\alpha = (\ell R)\ /\ D\ (1-R)$$

where R is the reflection coefficient at grain boundary and D is the grain size. Fig.(4) gives the relation between ρ and t for deposition rates equal 2 and 7 Å/S, where the theortical curve (continuous line) was calculated according to M-S theory. Values of ρ_o and $\ell_o(1-p)$ were taken as estimated from Fig.(3) and the grain size was taken equal to the thickness.

Two theoretical curves are shown in Fig.(4) with R = 0.05 and = 0.15 for comparison. For deposition rate equals 2 Å/S, the best fit was obtained when R=0.05. This small value of the reflection coefficient at grain boundary indicates that the contribution of the grain boundary scattering is not quite appreciable. This is not in agreement with previous data, namely R = 0.28(8) and R = 0.3(13). The higher R values assumed to be due to the higher deposition rate used by them, namely 10 Å/S. This statment was checked by studying the resistivity thickness relationship for deposition rate higher than 2 Å/S. Fig.(4) shows also ρ versus t for deposition rate equals 7 Å/S. The best fit gave $\rho_o = 2.53 \times 10^{-8}\ \Omega\text{m}$ and R = 0.15. The value of R obtained is in agreement with that found by Mayadas and Schatzkes(2) for Al-films but they did not specify the deposition rate. Thus the control of deposition rate is of vital importance.

Fig.(5) gives the variation of resistivity with thickness at normal and oblique incident at 60°. It can be shown that the angle of vapour incident greatly affected the resistivity of the film at all thicknesses. For deposition angles lower than 45°, the determined ρ/t curves were very close to that at normal incidence. The best fit to F-S theory gave $\rho_o = 14.9 \times 10^{-8}$ m and $\ell_o = 653$ Å at P = 0 when the angle of incidence equal 60°. Thus the theory is also applicable at oblique incident but with deposition angles higher than 45° the film resistivity is considerably increased.

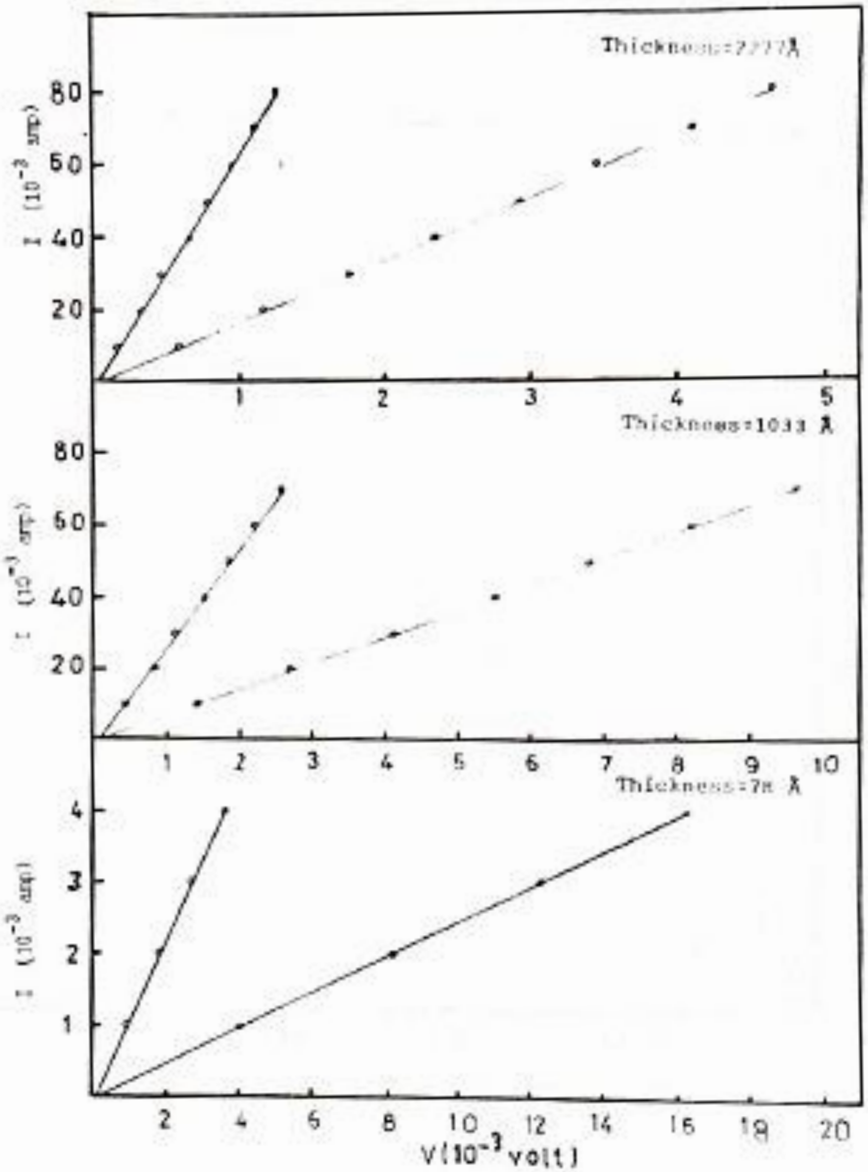

Fig.(1): Current-Voltage Characteristic for different film thickness.

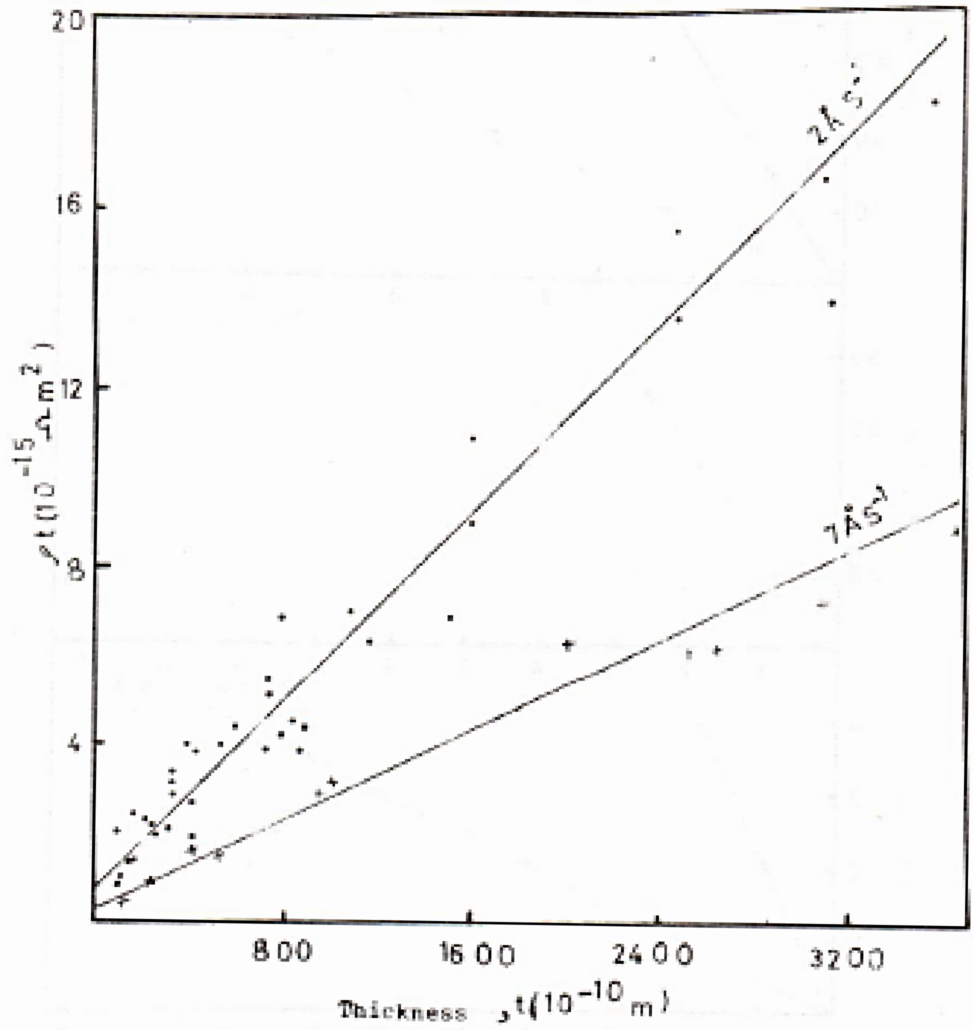

Fig.(2): Relation between ρt and t at deposition rates 2 and Å s^{-1}

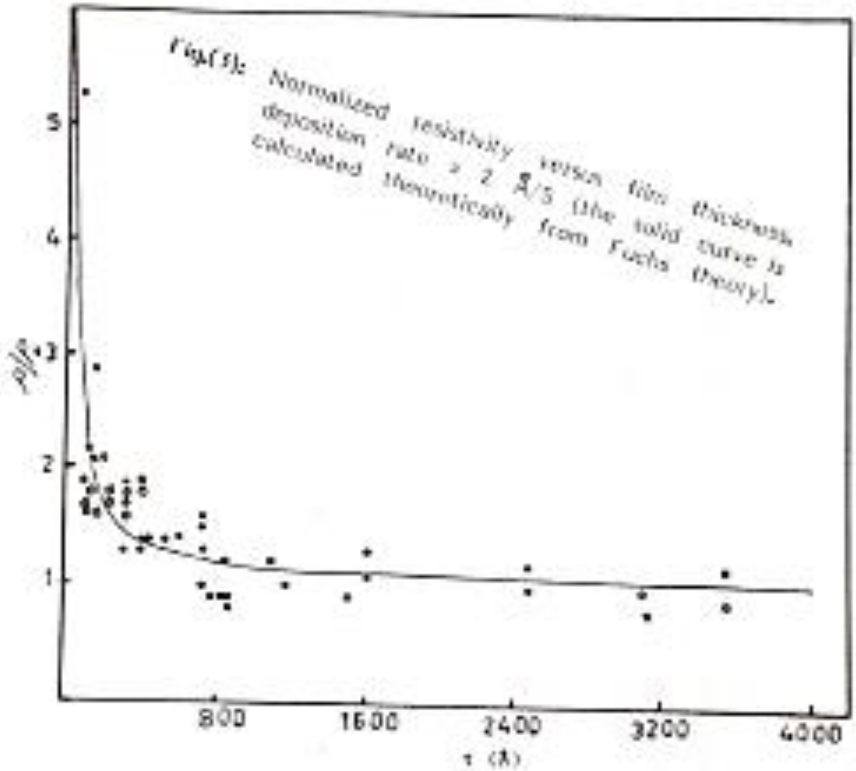

Fig.(5): Normalized resistivity versus film thickness deposition rate = 2 Å/S (the solid curve is calculated theoretically from Fuchs theory).

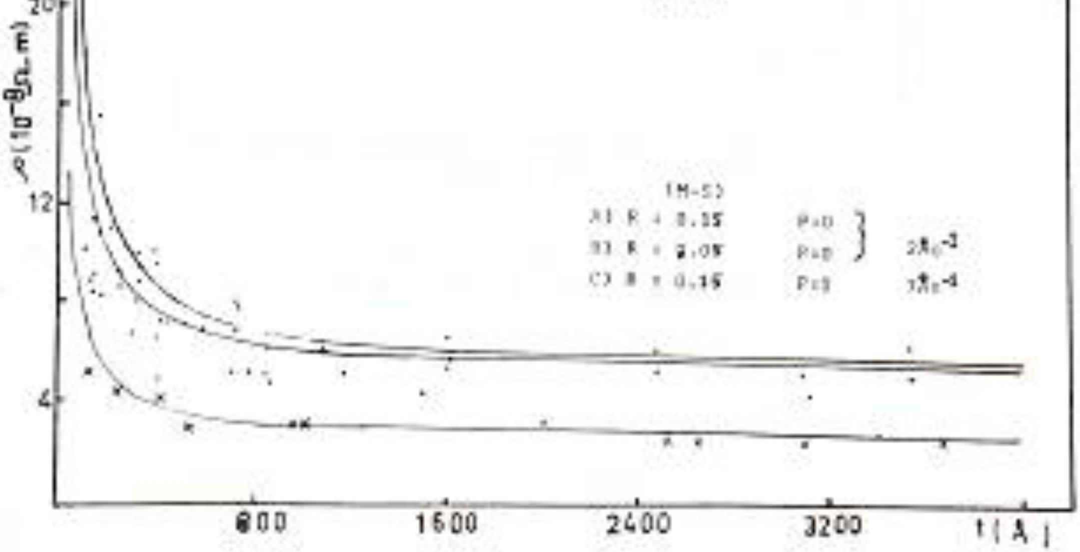

Fig.(4): Variation of resistivity with thickness measured at room temperature for deposition rates 2 & 7 Å S^{-1} (the solid curves are calculated theoretically from Mayadas theory).

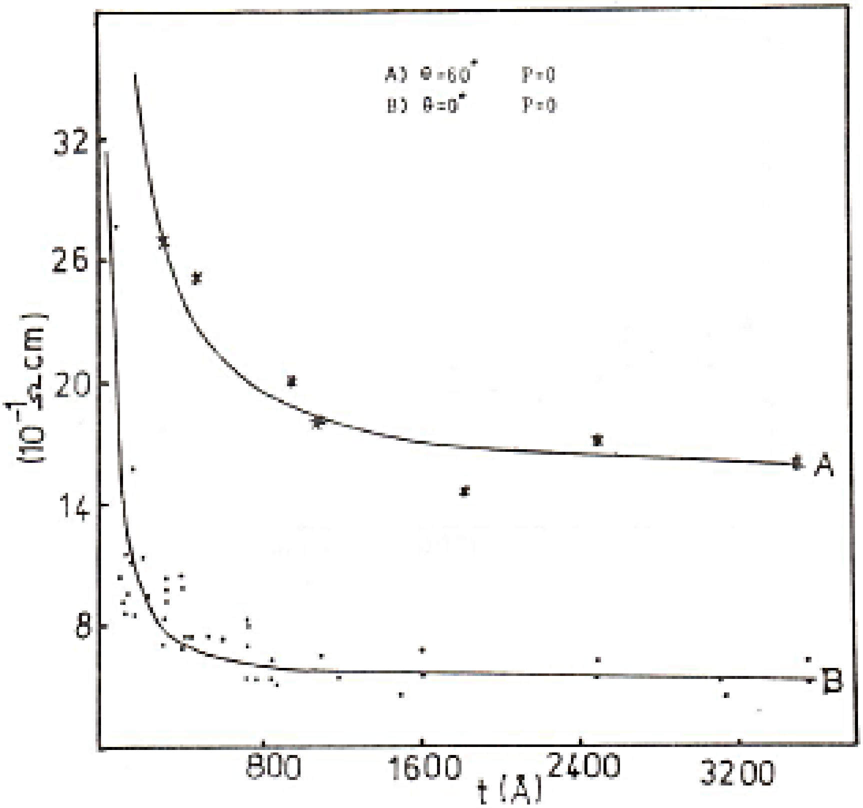

Fig.(5): Variation of resistivity with thickness at normal and oblique incidence (the solid curves are calculated theoretically from Fuchs theory).

Conclusions:

The here-reported measurements on the electric resistivity of polycrystalline Al-films show that:

1. Mayadas theory is generally applied fathfully than Fuchs one, with different values of R depending on the deposition condition.
2. The product $\rho_0 l_0$ is not a material constant but it depends on the deposition conditions.
3. Deposition rate affect both the total resistivity and the contribution of the grain boundary scattering.

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