THE EFFECT OF STRAIN ON THE ELECTRICAL PROPERTIES OF DISCONTINUOUS THIN METAL FILMS

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SUMMARY

The theory of the strain coefficient of resistance of discontinuous thin metal films is developed one stage beyond present levels by the inclusion of the variation of the activation energy. A variety of assumed island geometries is used in the development of simplified formulae. For all films tested, the activation energy decreases with strain giving a gauge factor which changes from positive at low resistance or high temperature to negative at high resistance or low temperature. This effect is predicted theoretically for flat disc-like islands where the strain is transmitted to the islands through points of adhesion with the substrate. In all cases the magnitudes of change observed are consistent with strains of approximately one order greater than those applied.

1. INTRODUCTION

It has been shown that the gauge factor, G of an ultra-thin metal film of resistance R is much greater than that of the bulk material, where G is defined for a film of length, L, as[1,2]

$$G = (\Delta R/R)/(\Delta L/L) \tag{1}$$

The increase in G is related to the discontinuous nature of such films which consist of a series of discrete metallic islands separated by small gaps. It has been shown that electrical conduction in such films proceeds by electron tunnelling through the substrate separating the islands[3,4]. In the charge transfer process the islands become electrostatically charged and this gives rise to an electrostatic activation

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energy, δE. The film resistance is expressible, for most practical purposes, as

$$R = Ad^2 . \exp (4\pi d/h)(2m^* \bar{\phi})^{1/2} . \exp \delta E/kT \tag{2}$$

where k is Boltzmann's constant, h is Planck's constant, d is the island separation, m^* is the effective mass of the tunnelling electron, $\bar{\phi}$ is the height of the tunnelling barrier, T is the absolute temperature and although A also depends on m^*, $\bar{\phi}$ and d, the sensitivity to variation of any of these is less than that of the exponential term and A may reasonably be considered constant.

As the film is extended it is expected that d will increase and film resistance will increase. All existing theories have been limited to this aspect which results in[5]

$$G = (4\pi L/h)(2m^* \bar{\phi}) \tag{3}$$

unless the pre-exponential dependence on d is included[6]. The image reduction[3] of $\bar{\phi}$, which also varies with d, has always been neglected.

The activation energy term δE is also a function of film geometry but has been neglected in the theoretical treatments. Preliminary tests, however, have indicated that δE varies substantially under stress[7]. An estimated strain, ε, of 5×10^{-4} ($\varepsilon = \Delta L/L$) gave a 5% reduction in δE (Fig. 1). The pre-exponential

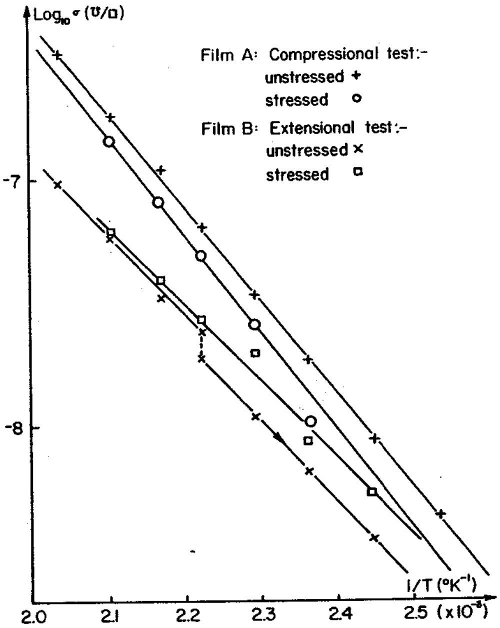


Fig. 1. Effect of compressional and extensional stress on two films.

resistance term increase in R was more than offset by the decrease in δE in these preliminary experiments resulting in a net decrease in R at room temperature with extensional strains. This result must be compared with the usual increase in film resistance with film extension.

The discrepancy suggested determining the conditions under which resistance decreases with extension. In this paper the contributions of both the tunnelling and activation terms to the gauge factor are determined theoretically for a variety of idealised film structures. The results of experimental determinations of the variations of both terms are compared with the theory.

2. THEORY

2.1. *Approximation of film structure*

Early analyses of the activation energy of conduction of discontinuous thin metal films assumed that the islands were hemispherical in shape and approximated this form by spherical islands. A variety of workers[8–10] have concluded, however, that the islands have a flattened shape which may be represented mathematically as an oblate spheroid of eccentricity, l. The oblate spheroid reduces to a sphere when $l = 0$ and to an infinitely thin disc for $l = 1$[7–10]. The oblate spheroid shape is clearly only exact for a substrate–island contact angle equal to π, *i.e.* for no substrate–island interaction. The binding of gold (which is used in the experimental part of this study) to glass is very weak and the approximation of oblate spheroid islands is probably best for this combination. Even here, however, the contact angle is less than π and the island is in direct contact with the substrate for some finite area rather than the single point implied by the true oblate spheroid form[8, 11].

2.2. *Variation of geometry with strain*

On the assumption of an oblate spheroid island shape there is only a point contact between the island and the substrate. No strain can therefore be transmitted from the substrate to the island and the latter retains its original shape and dimensions at all times. Under these circumstances (termed henceforth "no adhesion") only the island separation, d, varies while l and the major axis island radius, r, parallel to the substrate, remain constant. The separation $d(\varepsilon)$ is given by

$$d(\varepsilon) = d_0 + D_0\,\varepsilon \tag{4}$$

where d_0, r_0, D_0 are the unstrained values of d, r, D respectively and $D = 2r + d$.

For the case of finite bonding between the substrate and island (contact angle $< \pi$) the island dimensions and l will clearly change with substrate strain. In the extreme case of a disc shape in contact with the substrate at all points the parameter $p = d/D$ remains constant with applied strain and

$$D(\varepsilon) = D_0\,(1 + \varepsilon) \tag{5}$$

At this stage the approximations are made for the adhesion case that (i) D varies according to eqn. (5) for all oblate spheroid shapes (strictly true for $\pi/2$ contact angle only) and (ii) that the islands retain the form of an oblate spheroid of eccentricity l (strictly true for $l = 1$ only). For constant island volume

$$l(\varepsilon) = \{1-(1-l_0^2)/(1+\varepsilon)^6\}^{1/2} \tag{6}$$

where l_0 is the unstrained value of l. Any real case then is expected to lie between the conditions described by eqns. (4) and (5) with (6), and it is necessary to keep the approximations in mind when representing any real case.

2.3. *Variation of activation energy with strain*

In general, the activation energy for the oblate spheroid form is expressible (neglecting image effects[12]) as[7, 13, 14]

$$\delta E = (e^2/4\pi\varepsilon) r^{-1} l^{-1} \sin^{-1}[lr/(r+d)] \tag{7}$$

$$= (e^2/4\pi\varepsilon) D^{-1} l^{-1} 2(1-p)^{-1} \{\sin^{-1} l - \sin^{-1}[l(1-p)/(1+p)]\} \tag{8}$$

Substitution of eqns. (4), (5) and (6) in (7) and (8) where appropriate gives

$$\delta E(\varepsilon) = \delta E_0 + \frac{e^2}{4\pi\varepsilon}\cdot\frac{1}{l_0 r_0}\left[\sin^{-1}\frac{l_0 r_0}{r_0+d_0} - \sin^{-1}\left\{\frac{l_0 r_0}{r_0+d_0}\left[1+\left(1+\frac{r_0}{r_0+d_0}\right)\varepsilon\right]^{-1}\right\}\right] \tag{9}$$

for the no adhesion case, (r, l constant), and

$$\delta E(\varepsilon) = (e^2/4\pi\varepsilon) D_0^{-1} 2(1-p_0)^{-1} (1+\varepsilon)^{-1} l^{-1} \{\sin^{-1} l - \sin^{-1}[l(1-p_0)/(1+p_0)]\} \tag{10}$$

for the adhesion case (p constant) where δE_0, p_0 are the unstrained values of δE, p and l in eqn. (10) is given by eqn. (6).

In the particular cases of the sphere and disc[3, 7, 14]

$$\delta E_0 = (e^2/4\pi\varepsilon)(r_0^{-1} - (r_0+d_0)^{-1}) \tag{11}$$

$$\delta E_0 = (e^2/4\pi\varepsilon) D_0^{-1} 2(1-p_0)^{-1} \tan^{-1}[2p_0^{1/2}/(1-p_0)] \tag{12}$$

respectively,

$$\delta E(\varepsilon) = \delta E_0 + (e^2/4\pi\varepsilon) D_0^{-1}.4\varepsilon.(1+p)^{-1}(1+p+2\varepsilon)^{-1} \tag{13}$$

for sphere—no adhesion,

$$\delta E(\varepsilon) = (e^2/4\pi\varepsilon) r_0^{-1}.\tan^{-1}[D_0^{1/2}.(1+\varepsilon)^{1/2}.(d_0+D_0\varepsilon)^{1/2}.r_0^{-1}] \tag{14}$$

for disc—no adhesion, and

$$\delta E(\varepsilon) = \delta E_0/(1+\varepsilon) \tag{15}$$

for disc with adhesion. The final case of a sphere with adhesion is given by substituting $l=(1-(1+\varepsilon)^{-6})^{1/2}$ in eqn. (10).

A convenient form for presentation of the results is as $(\delta E(\varepsilon)-\delta E_0)/\delta E_0$ as a function of ε. For the no adhesion cases $(\delta E-\delta E_0)/\delta E_0$ is given by

$$\text{Sphere: } \frac{1-p}{1+p} \cdot p\varepsilon \cdot \left(1+\frac{2\varepsilon}{1+p}\right)^{-1} \approx \frac{1-p}{1+p} \cdot p\varepsilon \tag{16}$$

$$\text{Disc: } \frac{\tan^{-1}[2(1+\varepsilon)^{1/2}(p+\varepsilon)^{1/2}(1-p)^{-1}]}{\tan^{-1}[2p^{1/2}(1-p)^{-1}]}-1 \tag{17}$$

Oblate Spheroid:

$$\frac{\sin^{-1}[l_0(1-p)/(1+p)]-\sin^{-1}[l_0(1-p)/(1+p+2\varepsilon)]}{\sin^{-1}l_0-\sin^{-1}[l_0(1-p)/(1+p)]} \tag{18}$$

and for the adhesion cases by

$$\text{Disc: } -\varepsilon/(1+\varepsilon) \approx -\varepsilon$$

Oblate Spheroid:

$$\frac{l_0}{l(1+\varepsilon)} \cdot \frac{\sin^{-1}l-\sin^{-1}[l(1-p)/(1+p)]}{\sin^{-1}l_0-\sin^{-1}[l_0(1-p)/(1+p)]}-1 \tag{19}$$

where l is given by eqn. (6). The approximations are for ε very small and it will be seen from the theoretical plots (Figs. 2 and 3) that the variations are linear with ε for small ε.

2.4. *Variation of tunnelling term with strain*

The structure sensitive portion of the tunneling term is

$$\theta = Ad^2 \exp[(4\pi d/h)(2m^*\bar{\Phi})^{1/2}] \tag{20}$$

and, neglecting the complex variation of $\bar{\Phi}$ with d due to image effects[3]

$$\ln(\theta/\theta_0) = 2\ln(d/d_0) + (4\pi/h)(2m^*\bar{\Phi})^{1/2}(d-d_0) \tag{21}$$

where θ_0 is the unstrained value of θ. In the two cases under consideration now.

$$\ln(\theta/\theta_0) = 2\ln(1+\varepsilon) + (4\pi/h)(2m^*\bar{\Phi})^{1/2}\, d_0\varepsilon \tag{22}$$

for the adhesion case, and

$$\ln(\theta/\theta_0) = 2\ln(1+\varepsilon D_0/d_0) + (4\pi/h)(2m^*\bar{\Phi})^{1/2} D_0\varepsilon \tag{23}$$

for the case of no adhesion. In both cases

$$\frac{\partial \ln(\theta/\theta_0)}{\partial\varepsilon} \approx [2+(4/h)(2m^*\bar{\Phi})^{1/2}d_0]\,\gamma \tag{24}$$

for small ε, where $\gamma=1$ without adhesion and $\gamma=D_0/d_0$ with adhesion. The

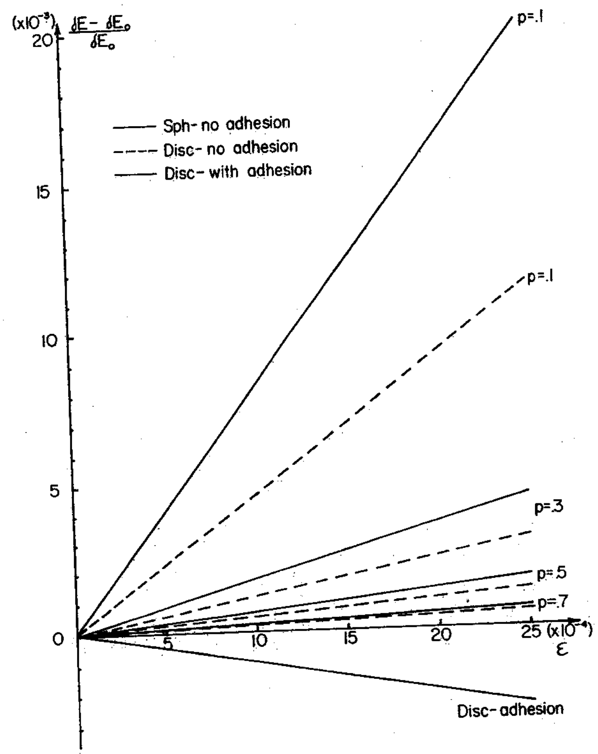


Fig. 2. Normalized variation of δE with strain for the extreme cases of disc-shaped and spherical islands.

second (exponential) term is dominant for typical films provided m^* is greater than approximately 10^{-3} times the electron rest mass.

2.5. *Substrate tunnelling*

In the foregoing expressions for δE (eqns. (7)–(19)), it has been assumed that the tunnelling electron leaves the island along an extension of the major axis. It has already been stated, however, that tunnelling takes place *via* the substrate which will modify the system electrostatic energy as the electron moves away from the island provided the contact angle is greater than $\pi/2$. This perturbation of the idealised case will be only briefly considered.

The modifications to conduction theory which are necessary when the true tunnelling path is considered have been described in a prior publication[15]. In the extreme case of point contact between the island and the substrate (contact angle $= \pi$) there is no adhesion and using the revised formulae for δE_0[15]

$$\delta E(\varepsilon) = (e^2/4\pi\varepsilon r_0) \tan^{-1}[(3+2\varepsilon-p_0)(1+2\varepsilon+p_0)/(2(1+\varepsilon)(1-p_0))] \tag{25}$$

for disc islands (*c.f.* $\delta E_0 = (e^2/4\pi\varepsilon r) \tan^{-1}[(3-p)(1+p)/2(1-p)]$)

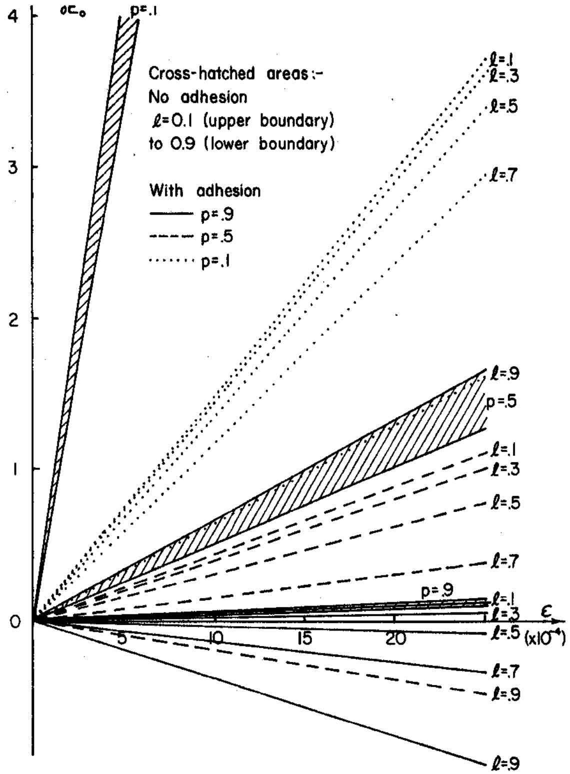


Fig. 3. Normalized variation of δE with strain for oblate spheroid islands.
(No adhesion case–cross-hatched areas)
(Adhesion case–individual lines)

and $\delta E(\varepsilon) = (e^2/4\pi\varepsilon D_0)\, 2[(1-p_0)^{-1} - (4(1+\varepsilon)^2 + (1-p_0)^2)^{-1/2}]$ (26)

for spherical islands

$$(c.f.\ \delta E_0 = (e^2/4\pi\varepsilon D)\, 2[(1-p)^{-1} - (4 + (1-p)^2)^{-1/2}])$$

The corrected normalised plots are presented in Fig. 4. It may be easily shown that the variation of the tunnelling term is given by either of eqns. (22) and (23) with the substitution of D_0 for d_0 wherever the latter appears.

2.6. *Island distortion*

If the islands genuinely approximate a section of the oblate spheroid in the unstrained condition then in a real case of a finite area of contact between substrate and island the form will distort under strain. The next stage then must be to determine the general expression for the distorted shape and evaluate δE for the revised form. This step has not yet been taken since the added complexity is not justified by present experimental accuracy.

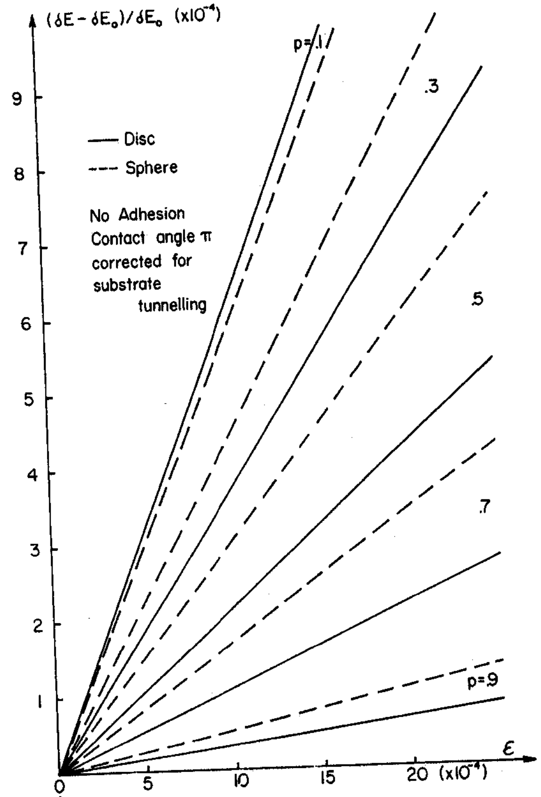


Fig. 4. Normalized variation of δE with strain for spherical and disc islands with π contact angle and corrected for substrate tunneling.

3. EXPERIMENTAL PROCEDURE

Discontinuous gold films with aluminum or gold contacts were used. The gold used is 99.99% pure, glass substrates were employed and the films were deposited in a variety of vacuum systems at around 10^{-6} τ. Film resistance was monitored during deposition with a Keithley electrometer which was also used for resistance measurements under strain testing. The substrate temperatures of films C, D and E below were maintained during deposition at greater than 200 °C to minimize annealing effects during subsequent thermal cycling. These three films were deposited on Corning 7059 glass. Films A, B, and F were (mil) deflection at the centre point with the ends held rigid. Stressing was performed inside an oven operated from ambient to about 200 °C. For substrate of length L and thickness $2t$ strained into an arc of radius $\bar{r}$ by a centre deflection h, the strain at the outside surface is given by[16]

$$\varepsilon = t/\bar{r}$$
$$= (2t/L) \cos^{-1}(1 - h/(\bar{r} - t))$$

and for very small deflections $h \ll L$

$$\varepsilon \approx 8ht/L^2$$

For a maximum of $h = 0.04$ in. and the measured substrate dimensions, $\varepsilon \gtrsim 2 \times 10^{-3}$.

Data for films C to F were obtained by decreasing ε as T fell slowly due to the hysteresis of the temperature control cycle. The significance of this measure will become apparent later.

The small resistance changes observed under stress made it impracticable to determine strain changes in the tunnelling and activation terms graphically. All data were, therefore, initially plotted to detect (i) any curvature of the Arhennius plots (in which case the characteristics are subdivided into approximately linear segments) (ii) any discontinuities indicative of structural changes during heating and (iii) isolated inconsistent measurements, and were then averaged analytically.

4. EXPERIMENTAL RESULTS

Witt and Coutts have referred to the difficulty of obtaining reliable values of the strain coefficient of resistance of discontinuous films due to stability problems[17]. Films A and B underwent rapid structural changes as the films were stressed and a continuous " flutter " in the resistance reading was experienced with films C and D. Films E and F showed no instability.

The results are summarized in Table I. θ_0 and δE_0 are found by plotting θ and δE *versus* ε to obtain a value for $\varepsilon = 0$ rather than by accepting the simple measured values (*e.g.* Figs. 5 and 6). A transmission electronmicrograph of film E is shown in Fig. 7. Films C and D are similar but with the larger islands generally linked together to form nearly continuous film structures. Films A, B and F are more filamentary with fewer islands scattered within the discontinuities. The activation energy of conduction is primarily dependent upon the smaller islands in the gaps separating the aggregates but dimensions are clearly distributed leading to curvature of the Arhennius plot for film E.

There are a number of significant features. (A) The scatter in the points of Figs. 5 and 6 is not exaggerated considering the stability problems encountered

TABLE I

EXPERIMENTAL RESULTS

Film	Vacuum System	θ_0 (Ω)	δE_0 (eV)	$\frac{\partial \ln(\theta/\theta_0)}{\partial \varepsilon}$	$\frac{\partial(\delta E - \delta E_0)/\delta E_0}{\partial \varepsilon}$	m^* (x rest mass)	G (at amb. temp)
A*	Leybold–Elliott oil diffusion			c.462	c. −104		NEG.
B*				c.673	c. −72		NEG.
C	NRC oil diffusion	3.17×10^6	0.2998	102	−4.23	1.71	+51
D		1.00×10^5	0.1353	227	−56	8.5	−73
E**	Varian ion system	8.32×10^6	0.2662	116	−13.8	2.2	−29
		1.05×10^8	0.1689	313	−74	16	−187
		5.01×10^8	0.1168	NEG***	+60		
F†	NRC oil	27.2	0.71	1800	−169		−3000

* Films destroyed before stripping for electron microscopy. Results analysed graphically only and are therefore approximate.
** Arhennius plot curved, approximated by three linear sections.
*** Decrease in θ with strain, not theoretically possible.
† Experimental results widely scattered, apparent anomalies disregarded.

with other films. (B) In all cases but one δE decreases with strain and for the single exception θ decreases. There is no apparent explanation for such a decrease in θ and this exceptional result will therefore be disregarded. (C) On the basis of the decreases in δE, it is initially concluded that the islands are flattened in form and adhere to the substrate. (D) According to the theory above the maximum possible negative value of $\partial[\delta E - \delta E_0)/\delta E_0]/\partial\varepsilon$ is -1 for disc islands (Fig. 2). The results cited in Table I give values up to two orders of magnitude greater. (E) Comparison of eqn. (24) with the results leads to the conclusion that the active portion for strain changes is the exponential. Assuming adhesion, m^* has been calculated for the films on Corning 7059 using an estimated $D_0 = 100$ Å and $\bar{\phi} = 0.59$ eV.[3]. (F) The values of m^* listed in Table I are all excessive (>1), m^* being expected to lie between 0.1 and 1 times the rest mass[3, 6, 7]. (G) From eqns. (1), (2) and (20) it can be shown that $G = \partial(\ln R)/\partial\varepsilon = \partial \ln(\theta/\theta_0)/\partial\varepsilon + (\delta E/kT).\partial((\delta E - \delta E_0)/\delta E_0)/\partial\varepsilon$ whence the values in Table I have been obtained. Note that G is positive at the actual measurement temperatures (greater than ambient) for films on Corning 7059 (films C, D and E) and that the transition to negative G occurs at very high resistance values (low temperatures). For the films on soda-lime (A, B and F), G is negative over the entire temperature range

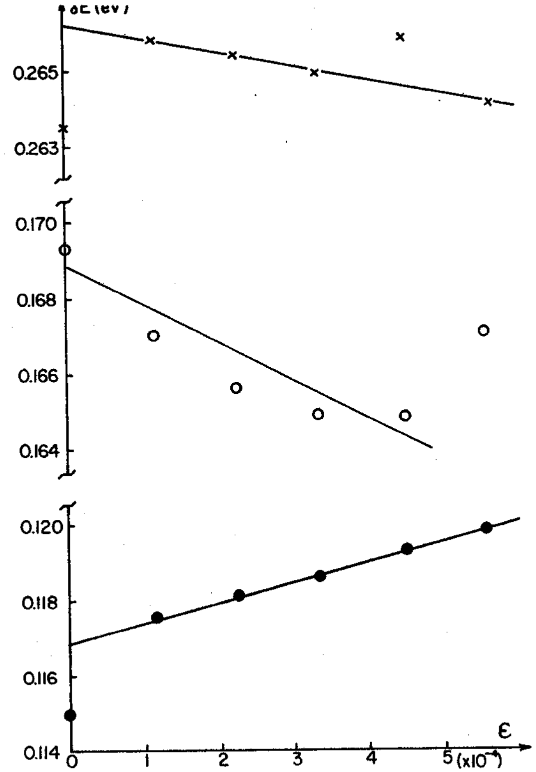


Fig. 5. Variations of δE with strain for film E.

5. DISCUSSION

There is a major discrepancy between the theory described and the experimental results. The increase in θ with ε is predicted for all models and the decrease in δE is consistent with the observation of flat islands[8] provided there is significant adhesion (*i.e.* contact angle $<\pi$). In the extreme case of a disc island in perfect contact with the substrate (contact angle of $\pi/2$) there is no correction necessary to account for substrate tunnelling and the modified formulae (Fig. 4) are inapplicable. This extreme case also leads to the maximum predicted negative value of $(\delta E - \delta E_0)/\delta E_0 = -\varepsilon$. The discrepancies between this figure (and expected values of m^*) and those actually obtained cannot be explained by error in the determination of the strain. Furthermore, while a real ellipsoidal island of large ℓ (nearly flat) will tend to flatten more with the distortion produced by stress and further reduce δE from its predicted reduction, the argument is not appropriate to the extreme approximation of a disc island now being employed. The influence of temperature drift during testing must, however, be considered.

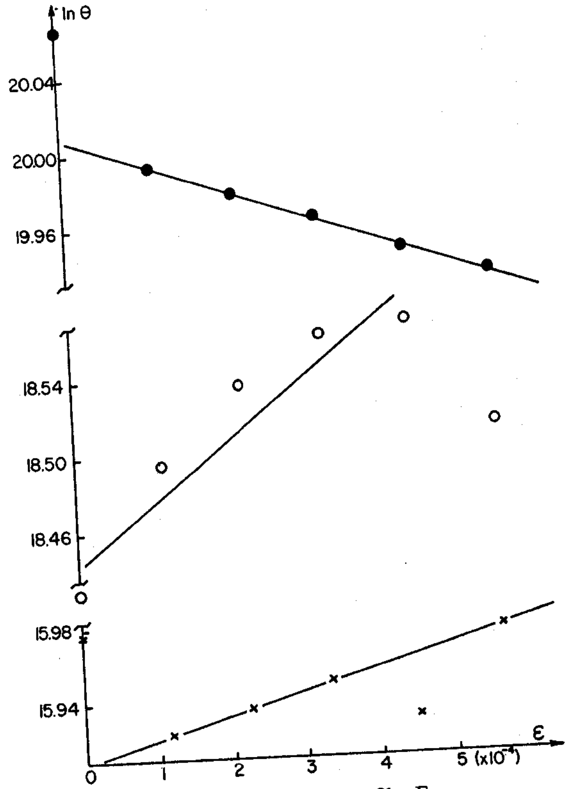
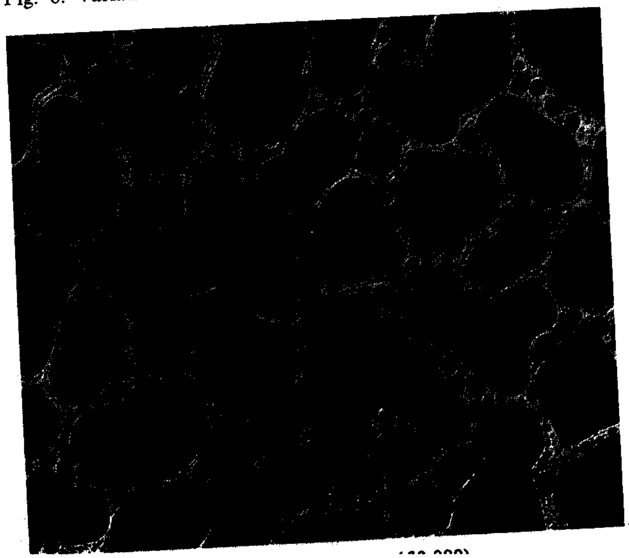


Fig. 6. Variation of θ with strain for film E.



It is clear from the magnitudes of change of R with stress shown in Fig. 1 that the reductions in δE are not entirely due to any spurious thermal drift effects which would be limited to a few degrees. For films C to F all testing was carried out at temperatures where the films displayed a positive value of G. As previously mentioned these tests were made by decreasing ε (decreasing R for positive G as observed) as temperature fell slightly due to the thermal control hysteresis (tending to increase R). This technique will result in a larger apparent reduction in δE and increase in θ than the true ones due to the higher rate of temperature fall at high temperatures, but the magnitudes involved (~ 1 °C) are unlikely to explain all the numerical anomaly.

6. CONCLUSIONS

Previous theoretical analyses of the strain coefficient of resistance of discontinuous metal films in terms of the tunnelling gap width alone have been shown to be inadequate for a full interpretation of the gauge factor since the activation energy is also strain dependent. The latter variation has been calculated for a variety of idealised island shapes and substrate–island interactions. Experiment indicates that δE decreases under strain which is in agreement with the theory for flattened islands with substrate adhesion. The magnitudes of the decrease in δE and increase in θ are up to two orders of magnitude greater than those predicted for adherent disc islands for which the maximum possible reduction in δE is predicted. No reasonable explanation for the discrepancy is offered.

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