NON-OHMIC PROPERTIES OF DISCONTINUOUS THIN METAL FILMS

J. E. MORRIS*

Department of Electrical Engineering, University of Saskatchewan, Saskatoon, Saskatchewan (Canada)

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SUMMARY

The electrostatically activated tunnelling model of conduction in discontinuous films is used to interpret non-ohmic effects. The usual spherical island model is extended to the cases of oblate spheroid and disc shaped islands, and to include the effects of island–substrate contact angle variation. Discrepancies between the full tunnelling theory and experiment are attributed to uncertainty in the determination of structural parameters. An interesting observation is the tendency to constant conductance at high fields with non-zero activation energy.

INTRODUCTION

The non-ohmic behaviour of discontinuous thin metal films[1–7] is well known and is generally attributed to a reduction of the activation energy of conduction according to the charge activated tunnelling model of conduction. However, with one possible exception[4] numerical agreement between published experiment and theory remains poor. The author's observations, in particular, are inadequately explained by the simplified treatments previously available. This paper is concerned with some modifications to the conduction theory and with a comparison of the full theory with experiment.

THEORY

Film conductance, σ, is generally expressed in the Arrhenius form

$$\sigma = \sigma_0 \exp \delta E/kT \qquad (1)$$

* Now at Physics Department, Victoria University of Wellington, P.O. Box 196, Wellington, New Zealand

where σ_0 is assumed to be constant, the activation energy δE decreases with applied field, k is Boltzmann's constant and T is the absolute temperature. According to Kiernan and Stops the full tunnelling expression is[8]

$$\sigma = \frac{a}{V}\cdot\frac{4\pi me}{h^3B^2}\cdot\frac{\pi BkT}{\sin\pi BkT}\exp - A\bar{\phi}^{1/2}\left(\frac{1-\exp - B(\delta E+eV)}{1-\exp(\delta E+eV)/kT} \pm \frac{1-\exp \pm B(\delta E-eV)}{1-\exp(\delta E-eV)/kT}\right) \tag{2}$$

where m is the electronic rest mass, e is the electronic charge, h is Planck's constant, $A=(4\pi d/h)\ (2m^*)^{1/2}$, m^* is the effective mass of the tunnelling electron, $B=A/2\bar{\phi}^{1/2}$, V is the voltage between metal islands, a is the effective electrode area, δE is the activation energy (considered in detail below), d is the inter-island gap width and $\bar{\phi}$ is the average barrier height between islands ($=\bar{\phi}_0(1-eV/2\bar{\phi}_0)$ where $\bar{\phi}_0=\bar{\phi}$ for zero field). σ_0 is clearly field dependent and this dependence must be included.

It has been shown for spherical islands[1–5], that for $0<E_a<E_{amin}$

$$\delta E = \frac{e^2}{4\pi\varepsilon R}\frac{4p}{1-p^2} - eRE_a \tag{3}$$

where $R=2r+d$, r is the island radius, d is the inter-island gap width, E_a is the applied field, $p=d/R$, ε is the effective dielectric constant, and

$$E_{amin} = (e/4\pi\varepsilon R^2)\,4p/(1+p)^2 \tag{4}$$

For $E_{amin}<E_a<E_{amax}$

$$\delta E = \frac{e^2}{4\pi\varepsilon R}\frac{2}{1-p} - 2\left(\frac{e^3E_a}{4\pi\varepsilon p}\right)^{1/2} + eRE_a\frac{1-p}{2p} \tag{5}$$

where

$$E_{amax} = (e/4\pi\varepsilon R^2)\,4p/(1-p)^2 \tag{6}$$

and for $E_a>E_{amax}$, $\delta E=0$.

The island shape is better approximated by an oblate spheroid[9–12] of eccentricity ℓ and major radius r. With this model[13–15]

$$\delta E = \frac{e^2}{4\pi\varepsilon R}\frac{1}{\ell}\frac{2}{1-p}\left(\sin^{-1}\ell - \sin^{-1}\frac{\ell(1-p)}{1+p}\right) - eRE_a \tag{7}$$

for $0<E_a<E_{amin}$,

$$\delta E = \frac{e^2}{4\pi\varepsilon R}\frac{1}{\ell}\frac{2}{1-p}\left(\sin^{-1}\ell - \sin^{-1}\frac{\ell}{1+2x/(1-p)R}\right) - eE_a\frac{x}{p} \tag{8}$$

for $E_{amin}<E_a<E_{amax}$, and $\delta E=0$ for $E_a>E_{amax}$, where

$$E_{amin} = (e/4\pi\varepsilon R^2)\,4p(1+p)^{-1}((1+p)^2-\ell^2(1-p)^2)^{-1/2} \tag{9}$$

$$E_{amax} = (e/4\pi\varepsilon R^2)\,4p(1-p)^{-2}(1-\ell^2)^{-1/2} \tag{10}$$

(11)

Substitution of $\ell = 1$ in eqns. (7) to (11) gives equivalent results for disc shaped islands.

A further modification is necessary (Fig. 1). The tunnelling distance has always been previously taken as the minimum gap width (*i.e.* path A, d). All

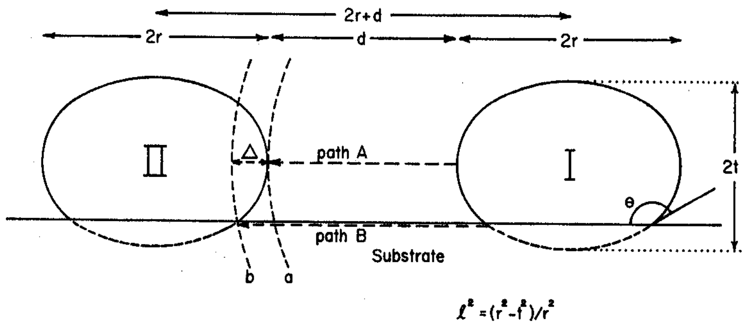

Fig. 1. Idealized film geometry for theoretical correction of δE for substrate tunnelling ($\theta = \pi/2$ – standard formulae, $\theta = \pi$ – modified formulae).

evidence, however, points to tunnelling *via* the substrate[7], (path B) and the prior approach is only valid for $\theta = \pi/2$. For noble metals on glass $\pi/2 < \theta < \pi$ ($\theta \approx 3\pi/4$ for gold[12, 16]) and the calculation of δE must be modified accordingly. For simplicity, only the extreme case of $\theta = \pi$ will be considered here. In general, for no field[13, 15],

$$\delta E = (e^2/4\pi\varepsilon\ell r)\,(\sin^{-1}\ell - \sin^{-1}(r\ell/(r+\lambda))) \tag{12}$$

where $r+\lambda (= r+d+\Delta)$ is the major axis of the oblate spheroidal equipotential surface b which is confocal with island I. For disc islands $r+\lambda = R$ and for spherical islands $r+\lambda = (R^2+r^2)^{1/2}$. An identical treatment to that employed above gives ($E_a < E_{a\,max}$)

$$\delta E = \frac{e^2}{4\pi\varepsilon r}\left(1 - \frac{1}{(1+(x/r)^2)^{1/2}}\right) - eE_a x \tag{13}$$

for spherical islands, and

$$\delta E = \frac{e^2}{4\pi\varepsilon r}\tan^{-1}\left[\left(\frac{x}{r}\right)^2 - 1\right]^{1/2} - eE_a x \tag{14}$$

for disc islands, where, for spherical islands

$x = R$ for $0 < E_a < E_{a\,\min}$

x is the real root of $(x^2+r^2)^{3/2} = x(e/4\pi\varepsilon E_a)$ for $E_{a\,\min} < E_a < E_{a\,max}$

$$E_{amin} = (e/4\pi\varepsilon)\, R(R^2 + r^2)^{-3/2} \tag{15}$$

and for disc islands

$$x = R \quad \text{for } 0 < E_a <_{amin}$$

$$x = (r/2^{1/2})\,(1 + [1 + (e/\pi\varepsilon E_a r^2)^2]^{1/2})^{1/2} \quad \text{for } E_{amin} < E_a < E_{amax}$$

$$E_{amin} = (e/4\pi\varepsilon)\, R^{-1}(R^2 - r^2)^{-1/2} \tag{16}$$

Again $\delta E = 0$ for $E_a > E_{amax}$ but now E_{amax} is best found iteratively since the electrostatic barrier shape is more complex due to the initial tunnelling parallel to the equipotential surfaces.

EXPERIMENTAL RESULTS

Gold films were deposited on glass substrates by filament evaporation at pressures between 10^{-6} and 10^{-5} torr. Non-ohmic effects were observed with seven films the properties of which are tabulated in Table I. All films appear

TABLE I

SUMMARY OF DEPOSITED FILM PROPERTIES

Film	2	3	5	6	7	14	W
*Substrate**	SL	SL	SL	SL	SL	SL	C
Deposition temperature (°C)	Amb	Amb	Amb	Amb	Amb	132	200
Av. thickness (Å)	60	100	44	75	62	290	272
Deposition rate (Å/min)	10.2	9	62	12.3	7.5	10.4	22.7
δE_0 (eV) Exp'tl.	0.84	0.79	0.79	0.87	1.05	0.82	0.32/0.09†
σ_0(℧ □) Exp'tl.	10	0.025	1.3	200	~1	0.067	$5.9 \times 10^{-8}/2.0 \times 10^{-10}$†
R(Å)	100	100	100	100	50	50	50
p	0.6	0.8	0.75	0.8	0.75	0.6	0.2

* SL – Soda-lime, C – Corning 7059
† Parallel conduction

filamentary. In films 14 and W the filaments are formed by coalescence of large aggregates and almost cover the substrate. In films 2 to 7 less than 50% of the substrate is so covered. Between the aggregates of films 14 and W there are many small islands with diameters 250 Å down to the resolution limit (~7 Å) of the electron microscope. The islands in films 2 to 7 are more dispersed, generally smaller and with a narrower distribution of sizes. Conduction across the discontinuities in the filaments clearly takes place *via* the secondary nucleation islands. Island diameters (Table I) have been determined as the most popular in the measured distributions and gap widths estimated as half the typical filament separation with weighting to low values.

The filamentary structures cause a field enhancement effect since the absence of any positive TCR or Hall effect shows that negligible voltage is dropped in the actual applied field and L is the effective filament length.

Non-ohmic conductance plots for films 7 and 14 are presented in Figs. 2 and 3 where the feature of interest is the levelling off of conductance at high fields

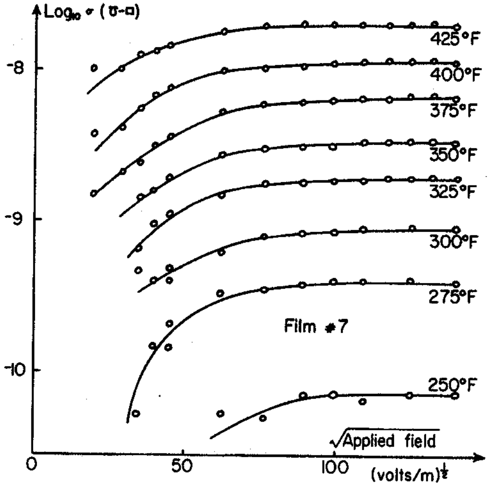

Fig. 2. Non-ohmic behaviour for film 7.

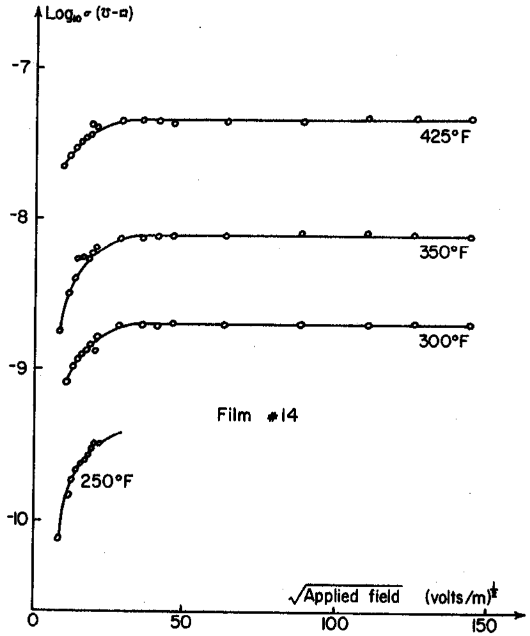

Fig. 3. Non-ohmic behaviour for film 14.

when δE is not reduced to zero. For films 2 and W, it cannot be determined whether the conductance variation is linear with E_a' or with $E_a'^{1/2}$, both giving reasonable agreement. Conductance varies approximately linearly with $E_a'^{1/2}$ for films 3, 5 and 6 but with some curvature at low fields similar to that in Figs. 2 and 3.

For film W, $L \approx 5000$ Å, $R \approx 50$ Å and the expected slope of log σ *versus* E_a' from eqn. (3) is 5.5×10^{-6} m/V which compares very favourably with the experimental figure of 3.8×10^{-6} m/V for $E_a' < 20 \times 10^3$ V/m $\ll E_{a\,min}' \approx 10^5$ V/m. Prior comparisons of theoretical δE with experiment suggest $\varepsilon_r \approx 2^{15}$ which is therefore used throughout this study.

The variation of δE with applied field is shown for four films in Fig. 4. Theoretical plots for the two extremes of island shape are also included and the points

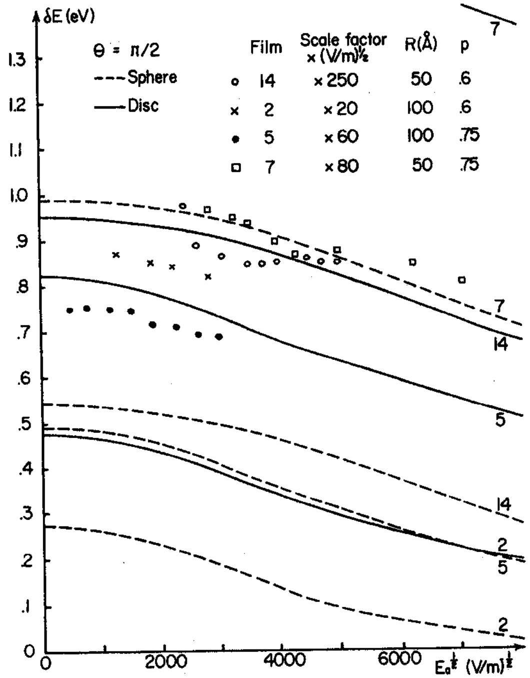

Fig. 4. Comparison of theoretical and experimental field reductions of δE.

have been fitted to these curves by use of the scale factors quoted. Derived values of (L/R) are reasonable for the filament dimensions in each case. Agreement is reasonable for the disc island approximation for films 5 and 14 and for the spherical approximation for film 7. Experiment is irreconcilable with theory for film 2.

The field variation of film conductance is superimposed on theoretical plots using the full tunnelling expression in Figs. 5 and 6. Vertical positionings of the experimental curves are arbitrary. The curves are compared with either the ... according to best agreement in Fig. 4 and the field enhanceagreement using measured film parameters except for film 2, for which no agreement was possible in Fig. 4.

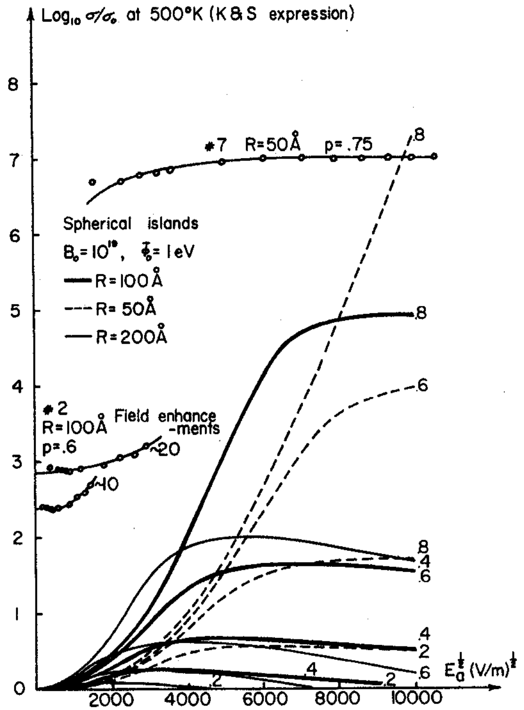

Fig. 5. Comparison of theoretical and experimental non-ohmic effect for spherical islands.

DISCUSSION

In the comparison of theory and experiment, only the case of $\theta = \pi/2$ was considered despite the development of a modified theory for $\theta = \pi$. A comparison of the two theories shows imperceptible differences for $p > 0.6$ in either δE or relative increases in conductance with field.

Equation (2) contains terms other than δE which are field dependent. The range of interest of E_a (Figs. 4 and 5) is up to 10^8 V/m *i.e.* up to $RE_a = 1$ for $R = 100$ Å. B increases at high fields as the barrier drops leading to reduction in $4\pi me/h^3 B^2$ which tends to zero when $eE_aR = 2\bar{\phi}_0$. It is this term which causes high field conductance decreases. The reduction in δE dominates all other non-ohmic effects in eqn. (2) provided it is assumed that m^* and d are small. No $\pi BkT/\sin(\pi BkT)$ curvature is observed in the Arrhenius plots of conductance. Therefore[7] $B < 10^{19}$ and for $\bar{\phi} \approx 1$eV, $d \approx 80$ Å it follows that $m^* \approx 10^{-3}\ m_e$.

Equation (2) may be expressed as

$$\sigma = \frac{a}{V}\frac{8\pi me}{h^3B^2}\frac{\pi BkT}{\sin \pi BkT}\exp - A\bar{\phi}^{1/2} \sinh \frac{eV}{kT}\exp - \frac{\delta E}{kT} \tag{17}$$

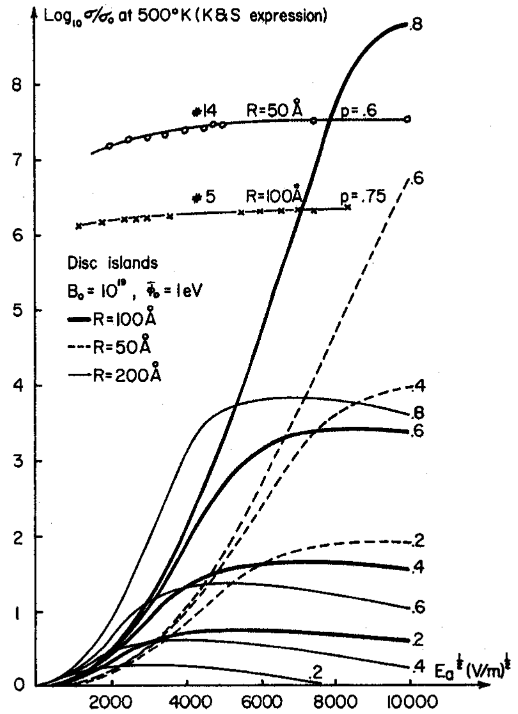

Fig. 6. Comparison of theoretical and experimental non-ohmic effect for disc islands.

for $eV \ll kT \ll \delta E$. The approximation may not be used for evaluation of non-ohmic effects here since eV is comparable with δE at the fields of interest due to the enhancement effects.

In Figs. 5 and 6 it is apparent that agreement would be better if effective p for each of the films investigated were much lower than the estimated values. The film structures are obviously highly irregular and clearly conduction will occur preferentially across gaps of minimum d, *i.e.* minimum p for constant R. It is worth noting then, that r is determined from observation of several hundred islands for film 14 only and that at the other extreme r was estimated for film 2 from the only five islands which could be located. In all cases it is reasonable to expect the estimation of R to be critical and lower values than those quoted may well be more correct. In this case p would be less than the present estimates. (Serious inaccuracy in the estimation of R for films with meandering filaments—2 to 7—is probable because of the large variation noted in the aggregate spacings.)

For smaller values of p, the reduction in theoretical δE will be largely offset by the increase caused by contact angles greater than $\pi/2$. In conclusion, then, the discrepancy between theory and experiment is wholly attributable to inadequate methods of determination of effective structural parameters.

CONCLUSIONS

The theory of non-ohmic behaviour in discontinuous metal films has been extended (a) to the generalised oblate spheroid island shape and flat disc islands, (b) to the case of a contact angle π between island and substrate, (c) to include the field dependence of the complete tunnelling expression. Film structures are highly agglomerated leading to field enhancement. A trend to constant conductance at high field, where δE is not near zero, has been noted and is predicted theoretically for certain film structures. Numerical comparison of prediction and theory for measured film geometries is poor but the problem lies more with the accuracy of determination of effective structural parameters than with the theory.

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