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A.C. EFFECTS IN ASYMMETRIC DISCONTINUOUS METAL FILMS

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Although the thermally activated electrical conduction mechanism in discontinuous metal island films on insulating substrates has been studied for many years, it still defies resolution at a detailed quantitative level. There is widespread (but not unanimous) agreement that the basic mechanism is by interisland tunneling, that the activation energy has electrostatic origins, and that percolation processes must be involved, although no simple percolation formalism has yet passed external criticism. The observation of strongly polarity-dependent resistances in asymmetrically deposited films suggests that the contact regions dominate film properties and a.c. measurements were performed to verify this model. The results support the concept fully, but at extreme asymmetries a pseudo-inductive effect develops, which leads to an extension of the basic model to include time-dependent space charge effects. With independent verification, this would be the first advance in the basic conduction model in over ten years.

1. INTRODUCTION

Discontinuous metal films are most readily formed in the initial stages of deposition and growth of a noble metal on an insulating substrate. The poor chemical bond between impinging atoms and the substrate surface leads to high surface mobility and to film formation as discrete metal islands instead of as a monolayer. Islands are typically 1–10 nm in diameter, separated by gaps of around 2 nm. For room temperature deposition, the discontinuous nature of the film persists to an average thickness around 5 nm [1].

Electrical conduction through the films is thermally activated and non-ohmic. The mechanism is generally thought to be due to electron tunneling between islands. The activation energy is due to the finite electrostatic energy necessary to charge an initially neutral island; it controls the effective carrier density by limiting the density of charged islands[2,3].

Most variations of the basic model, in calculating the activation energy, consider carrier generation (island charging by charge separation) to take place throughout the film[3,6]. It has been suggested, however, that the electrode region

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should control island charging since the energy required to charge a single island is only half that needed to create two oppositely charged islands in the bulk[7]. This view has been supported by the observation of asymmetrical electrical characteristics in films deposited off-center from the source so that the island structures immediately adjacent to the two electrodes are different[8].

Obviously, in a practical case there will be preferred paths through the film which may change as temperature or applied bias is varied. There have been many attempts to apply modified forms of percolation theory to the discontinuous film structure[3,6,9–11] but none has so far withstood independent critical scrutiny. The basic problem is that, to apply a simple percolation model to the entire film, one must assume some sort of correlation between the activation (charging) energy and tunneling distance (or gap width). The various percolation treatments all assume some such correlation, but none has ever been observed in practice[12]. If one considers the many variables available to a relatively simple ellipsoidal island model (three axes of rotation, random relative orientations in addition to sizes and separations) the null result is not surprising. Further complicating the issue is a lack of agreement on the power law relationship between the logarithm of log conductivity and inverse temperature in such films, so there can, similarly, be no agreement on what would constitute a successful theory.

It is the present author's contention that there are more fundamental issues to be pursued in the understanding of the conduction process before percolation concepts can be profitably added. One must understand the mechanisms in an idealized model with confidence before extrapolating to the real case. These mechanisms are not yet well understood[3,13].

For these reasons, the observations referred to earlier[8], of diode-like asymmetrical *I–V* characteristics in films with dissimilar island geometries adjacent to the electrodes, are considered to be so significant because the results are inconsistent with models based on equilibrium charged island densities in the bulk of the film, supporting, instead, early proposals of an electrode injection mechanism[7].

In this paper some preliminary results in the study of small signal a.c. impedances of asymmetrically deposited discontinuous metal films are reported. The first result of significance is the expected result in support of the electrode injection model, but a second observation at gross asymmetries suggest a further extension.

2. A.C. IMPEDANCE

For the purposes of discussion, an idealized discontinuous film structure is shown in Fig. 1. In the traditional view, the overall square film resistance is representative of that of a single island–gap–island combination within the film, leading to a single simple R_g–C_g equivalent circuit for both the film and the single gap (neglecting the small metallic series resistance R_b of the island itself). In this picture, R_b, R_g and C_g can each be determined from a.c. impedance measurements.

For an activation energy $\delta E \approx q^2/2C_g$ in the range 0.1–1 eV, C_g is expected to be of the order of 10^{-8}–10^{-7} pF. In practice, measured film capacitances as determined from Bode plot corner frequencies are typically around 1 pF[14–19]. There is a clear

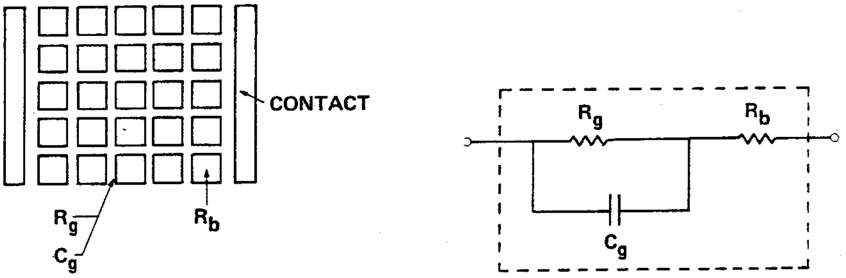

Fig. 1. Idealized discontinuous metal film structure: islands and gaps, square film.

inconsistency here which needs to be resolved[3]. It is noted, however, that the ratio of 10^7–10^8 is of the same order as the number of 1–10 nm islands (or parallel C_g elements) across a 1 cm film contact.

Films 0.125 in long by 0.75 in wide were deposited on Corning 7059 glass at room temperature by thermal evaporation from an alumina-coated tungsten boat in an oil diffusion vacuum system at a base pressure of 2×10^{-6} Torr. Asymmetric island structures were achieved by simultaneously depositing several films at varied angles of incidence from the vapor source (Fig. 2(a)). Film 20A was positioned

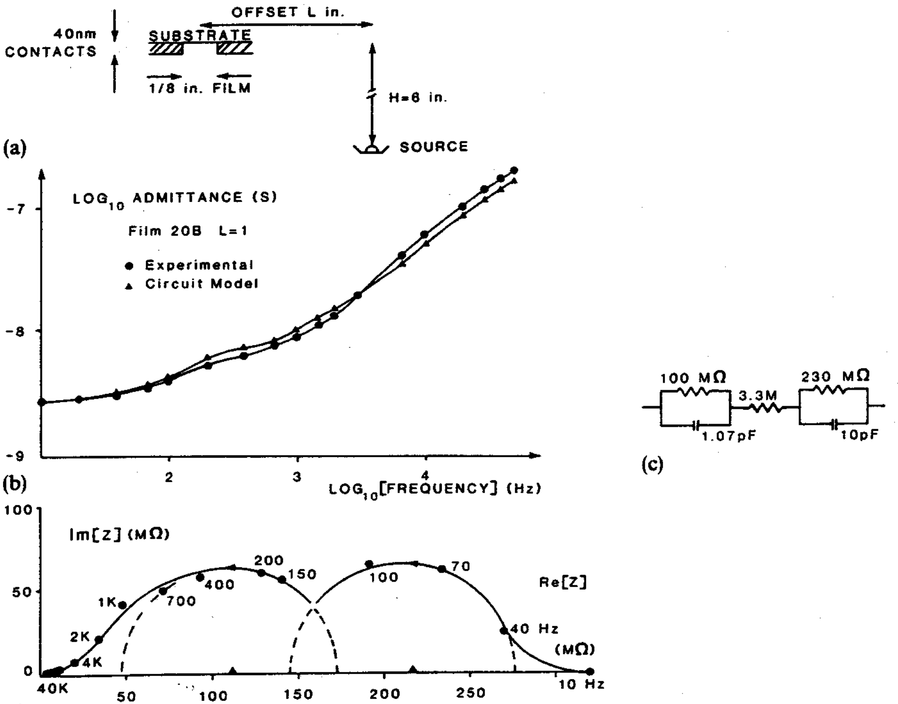

Fig. 2. (a) Deposition geometry for offset films. (b) Typical a.c. characteristics for small offset; note the double corner frequencies. (c) Circuit model for film 20B; the corresponding admittance plot is shown in (b). (d) Cole–Cole plot for films 20B; the semicircles shown with marked centers do not correspond to the circuit model in (c).

directly above the source; 20B (Figs. 2(b)–2(d)) was offset at about 5° from vertical incidence. 40 nm gold contacts were predeposited. The shadowing effect requires surface diffusion of the islands to establish a conduction path adjacent to the electrode[20] and effectively "thins" the film here as a result. A.c. measurements were made using a Princeton Applied Research PAR-122 lock-in amplifier, over its frequency range from 5 Hz to 50 kHz, with an internally generated 50 mV test signal.

It is important to establish a context for these results among others in the literature, especially since the system base pressure is not up to state of the art. Figure 3 shows the variation in d.c. activation energy with film thickness for normally deposited films in the context of other published data[21–25]. The correlation with data from Kazmerski and Racine for films deposited at a lower pressure (base pressure, 10^{-10} Torr)[23] is good, but the main point is that one can infer a similarly well-defined island structure, probably with a similar island size distribution double peaked at diameters around 2 nm and 20 nm with separations of more than 3 nm[23]. The films are clearly well away from the percolation threshold, despite Andersson's observation of an onset of continuity at thickness A (5.3 nm)[25].

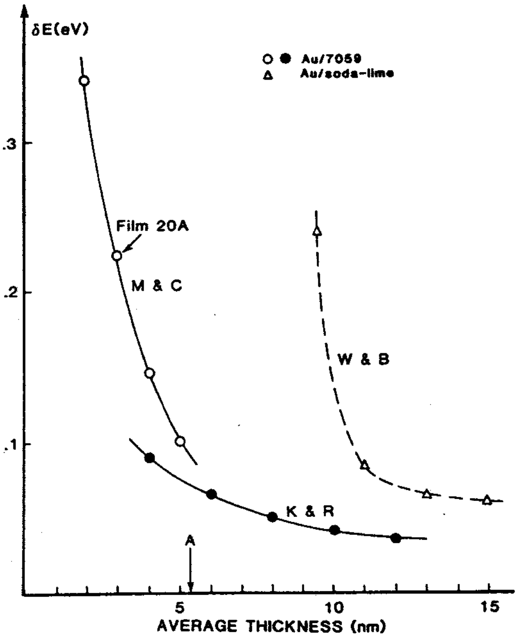

Fig. 3. D.c. activation energy of conduction and film thickness: M & C, Morris and Chiu[21,22]; K & R, Kazmerski and Racine[23]; W & B, Weitzenkamp and Bashara[24]; film 20A was codeposited with 20B (Fig. 2) at normal incidence; A, Andersson's[25] onset of continuity for gold on Corning 7059.

The basic interpretation of the results of Fig. 2 is clear; the parallel R–C combinations represent the contact regions of the film and the lower resistance series element corresponds to the bulk. Figure 2(c) emphasizes the deviations which still exist between this model and experiment, particularly at low and high frequencies. It is worth noting in passing that the roughly 3 MΩ "bulk" resistances of films 20A (not shown) and 20B exceed the maximum metallic resistance of about 40 kΩ/□ [26]. Even if this resistance is due to tunneling between islands in the film "bulk", past experiments show that it is not activated[14].

For the film 20A with $\delta E \approx 0.2\,\mathrm{eV}$ and $R_g = 2 \times 10^8\,\Omega$, C_g was measured at 3.4 pF, about 8×10^6 times $q^2/2\,\delta E$. For C_g to consist of 8×10^6 electrode–island capacitances along the contact, each would be about 2.5 nm wide, about the right order. Note that the corresponding electrode–island resistance of $1.6 \times 10^{15}\,\Omega$ may seem high, but is very reasonable when the tiny tunneling area of an individual island is taken into account.

In conclusion, a.c. impedance measurements on discontinuous thin metal films support but do not necessarily prove the concept of electrode carrier injection. The model has only been outlined in general terms, more confirmatory experimental work being required to clarify some remaining questions. It is, however, consistent with observations of asymmetric film conductances and resolves the inconsistency between film capacitance and activation energy which faults the elementary electrostatically activated tunneling model.

3. EXTREME ASYMMETRY

In Fig. 4, admittances are plotted for six simultaneously deposited films at increasing offsets L from the vertical. The unexpected "pseudo-inductive" effect at

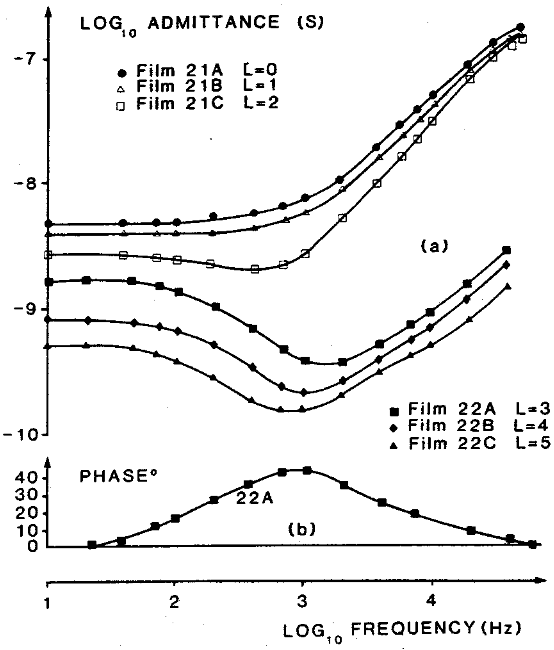

Fig. 4. A.c. admittance characteristics for six simultaneously deposited films with increasing offsets. (a) For 21A and 21B there is negligible offset effect, but for 21C the pseudo-inductive effect masks any development of a second R–C corner frequency. In films 22, the pseudo-inductive effect conceals the complete R–C roll-off. (b) Inductive phase shift for film 22A.

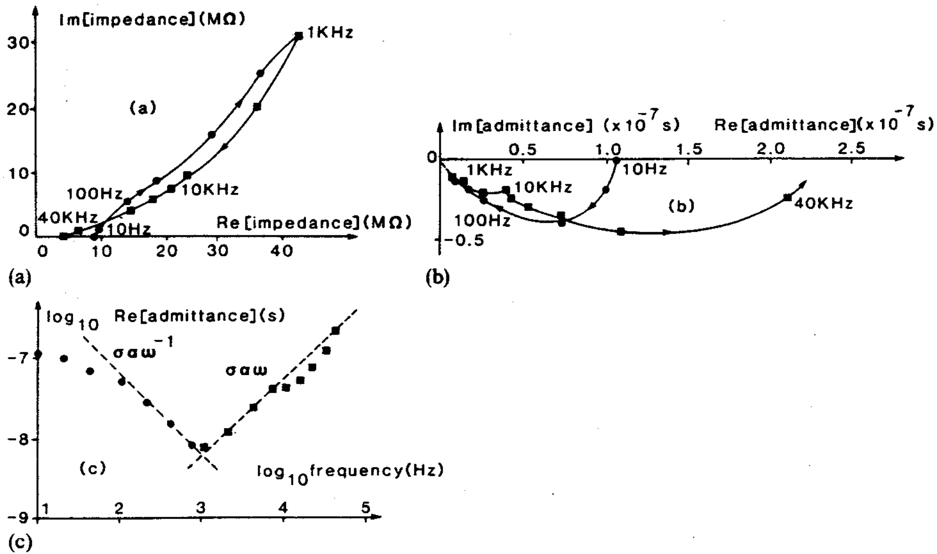

Fig. 5. Analysis of film 22A: complex plane plots of (a) impedance and (b) admittance with test frequency as the third parameter. (b) shows the pseudo-inductive effect at low frequencies and that conductance G increases with frequency at higher frequencies. (c) shows that the latter dependence of G on frequency is linear.

low frequencies is indicated by the initially decreasing admittance with frequency. Data for film 22A are replotted in various ways in Fig. 5 in an attempt to recognize some simple relationship in the data, but no single model emerges. It is tempting to write off the high frequency effect (Fig. 5(c)) as electron hopping on the basis of the linear variation in (real) conductance with frequency[27–29], but more systematic data are needed.

At this point, a low frequency model is being developed in which the relatively small bulk resistance R_s requires the injection of non-equilibrium excess charges at the electrodes. If the (d.c.) electrode injection impedance is sufficiently large, then one cycle may not provide sufficient time to inject the required carriers and admittance decreases until a more efficient alternative mechanism (capacitive or electron hopping) can be established.

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