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Modelling conduction in asymmetrical discontinuous metal thin films

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Abstract

The explanation of diode-like properties and pseudo-inductance in asymmetrical discontinuous metal films requires the addition of contact injection of charge carriers to the basic theory of charge activated tunnelling. Monte Carlo computer simulations of the conduction process demonstrate the observed phenomena, and the modified model can be used to gain greater insight into island film applications. © 1998 Elsevier Science S.A.

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1. Introduction

In the physical vapor deposition of a metal film on an insulating substrate, the film initially develops as a collection of discrete metal islands or nuclei. Significant electronic conduction is observed in discontinuous films with island diameters and gaps in the 1 to 10 nm range [1–4]. The electrical conductance may be expressed as

$$\sigma = \sigma_0 \exp - \delta E/kT,$$

where

$$\delta E = \left(q^2/4\pi\epsilon_r \varepsilon_0\right)\left(r^{-1} - (r+s)^{-1}\right)$$

is the zero field electrostatic charging energy of a small spherical island of radius r and separation s in a medium of relative dielectric constant ε_r, ε_0 is the dielectric constant of free space, q is the electronic charge, k is Boltzmann's constant, T is the absolute temperature, and

$$\sigma_0 = \lambda\left(4\pi m q^2/h^3 B\right)\cdot(\pi BkT)/\sin(\pi BkT) \cdot \exp - \left(A\phi^{1/2}\right),$$

with

$$B = 1/2\,A\phi^{1/2},\ A = 4\pi s(2m)^{1/2}/h,$$

is an electron tunnelling term for a square film of uniform islands, where h is Planck's constant, and λ, m, and ϕ are the effective tunnelling area, electronic mass, and tunnelling barrier height. Agreement between experimental and theoretical δE is sufficient to validate the electrostatic model [1,2], but absolute conductances are found to be orders of magnitude greater than theory.

Borziak et al. [5] reported three significant experiments. With previously deposited electrodes, they were able to fabricate discontinuous films with different island structures immediately adjacent to the contacts, with smaller islands and/or wider gaps. These symmetrical 'inhomogeneous' films showed that the voltage drop is always greater at the positive end of the film. The second result was the observation of stable and reproducible switching in such films, but the explanation of this effect is a goal of future work. In the third experiment, the inhomogeneous films were also made asymmetric, i.e. with different inhomogeneous structures at the two electrodes, whereupon the DC resistance became polarity dependent, i.e. a diode-like effect. These results cannot be explained on the basis of existing conduction models, and indicate that the conduction mechanism must depend significantly upon the islands at the electrodes [6,7].

The asymmetric film study has been extended to AC effects [8]. In the traditional model, the film is regarded as a matrix of identical island/gap elements, with the metal island resistance in series with the parallel combination of gap tunnel resistance R_g and capacitance C_g, where $\delta E = 1/2\,q^2/C_g$. C_g values determined by AC measurements on this model are universally orders of magnitude greater than those consistent with δE. With the asymmetrical inhomogeneous film, two corner frequencies appear, yielding two distinct values for both R_g and C_g, corresponding to the two electrodes. In addition, the C_g values match well to capacitances between the electrodes and film across a single gap width. At extreme asymmetries, a 'pseudo-in-

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ductive' effect makes an appearance, (as one contact resistance becomes very large,) representing a time delay to establish steady-state conductance in the film by charge carrier injection.

2. Modifications to the conduction theory

The results and arguments above lead to the concept of contact injection as the primary source of electrons and holes in the film, with the term 'hole' referring to a positively charged island. With zero field, bulk charge separation occurs by tunnelling between initially neutral islands, but immediate recombination will commonly follow. Eventually enough charges will drift apart to establish an equilibrium balance between generation and recombination, determined by the Boltzmann distribution. This is the traditional carrier density model which predicts lower conductances than observed. When a voltage is applied to the film, these carriers will begin to drift in the field, and additional carriers will be injected at the electrodes. As the injected carriers drift into the center of the film, the film conductance rises after some time to the steady state value. This is the origin of the pseudo-inductive effect, with the applied field governing the rate of current rise.

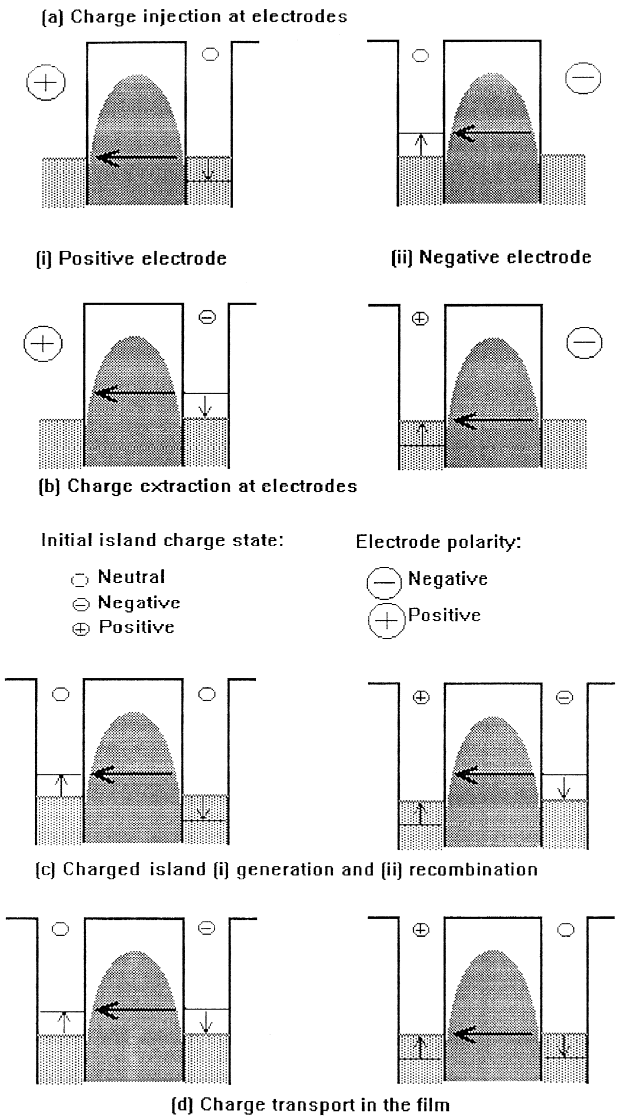

Fig. 1. Energy diagrams for electron tunneling. (a) Charge injection (initially uncharged islands), and (b) charge removal (initially charged islands), at (i) positive and (ii) negative electrodes. (c) (i) Generation and (ii) recombination, of charge carriers, and (d) charge transport in the central film.

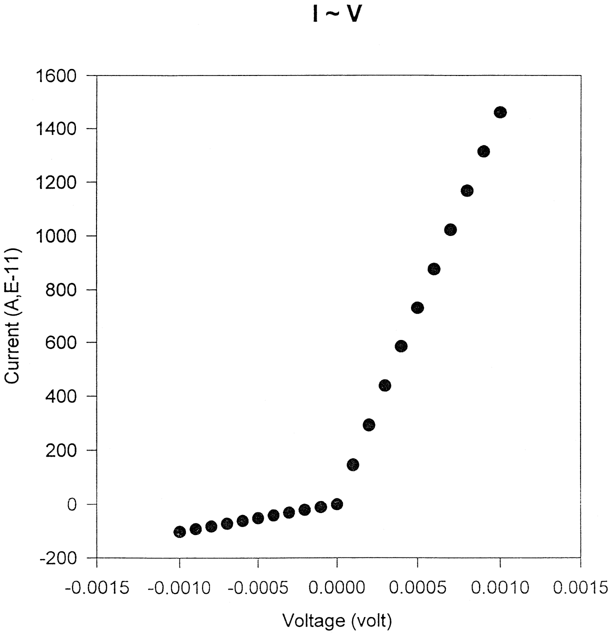

Fig. 2. Diode effect. (Compare figure 3a Ref. [5].)

The effects above apply to symmetrical film structures, whether homogeneous or inhomogeneous. If one adds the asymmetrical provision, then the different injection probabilities at the positive and negative electrodes produce the diode effect. The 'diode' effect requires a quantitative difference in the electronic transfer probabilities at the positive and negative electrodes. The basis of the difference is shown in Fig. 1, in which energy diagrams are presented for tunnelling transfers required for (a) charge injection and (b) removal, for (c) carrier generation and recombination, and for (d) charge transport in the central film. The quadratic tunnelling barriers shown include image charge effects. The shaded areas mark equilibrium Fermi levels, and the arrows indicate the δE electrostatic energy level shifts from initial to final Fermi levels in the islands. Electrode Fermi levels are fixed. The electrostatically charge activated injection processes will be the limiting factors in charge transport, and it is immediately apparent that injection tunnelling resistance at the negative electrode is less than that at the positive one.

A computer program has been written to model the electrical behavior of an idealized discontinuous thin film, to verify the concepts developed above by demonstration

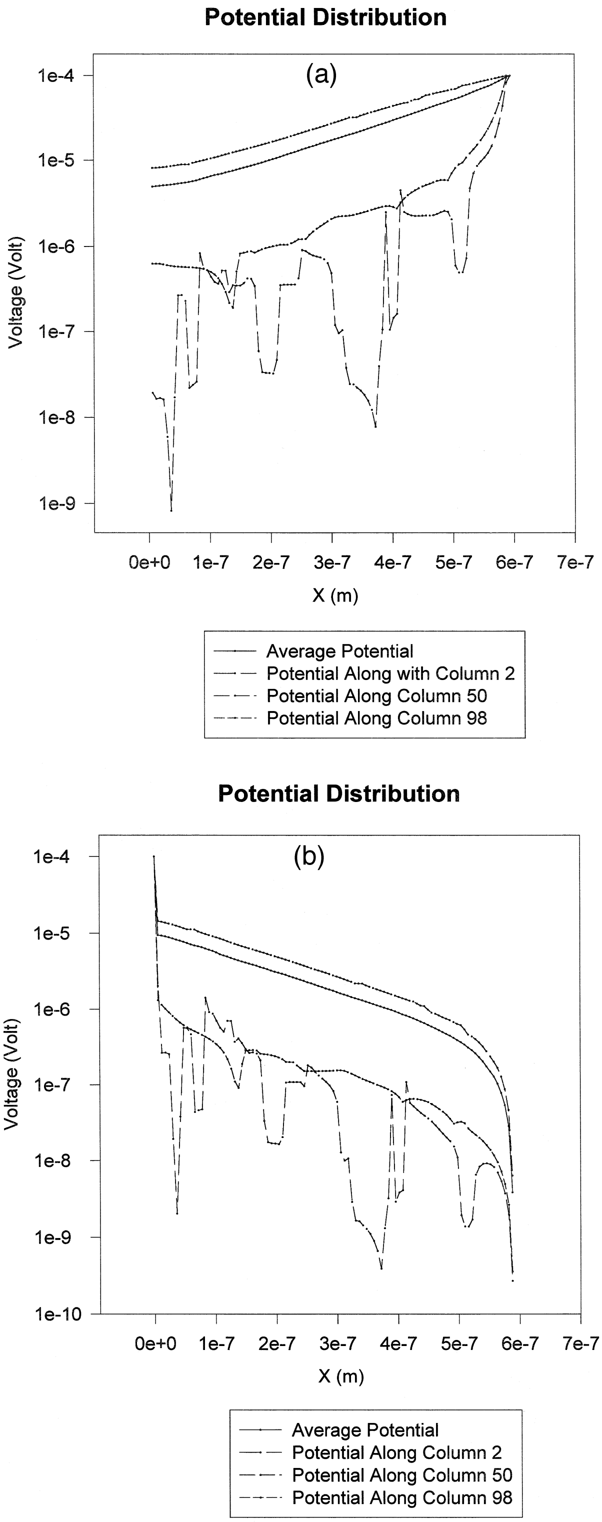

Fig. 3. Potential distributions along the film. (a) Forward/positive and (b) reverse/negative bias. (Compare figures 2c and b respectively, Ref. [5]). (i) Average across 100 columns; (ii) column 2; (iii) column 50 (central); (iv) column 98 (3rd from edge).

of the experimental effects described. The results described below are for a 100 × 100 array of metal islands on a regular square grid. With the exception of the 'anomalous' row of 1 nm islands 5 nm from the grounded electrode, all are 3 nm diameter islands separated by 3 nm gaps. The islands are assumed to be spherical for activation energy calculations, but cubic for the determination of tunnelling areas. $T = 300$ K, $\varepsilon_r = 2.5$, and $\phi = 1$ eV. The voltage

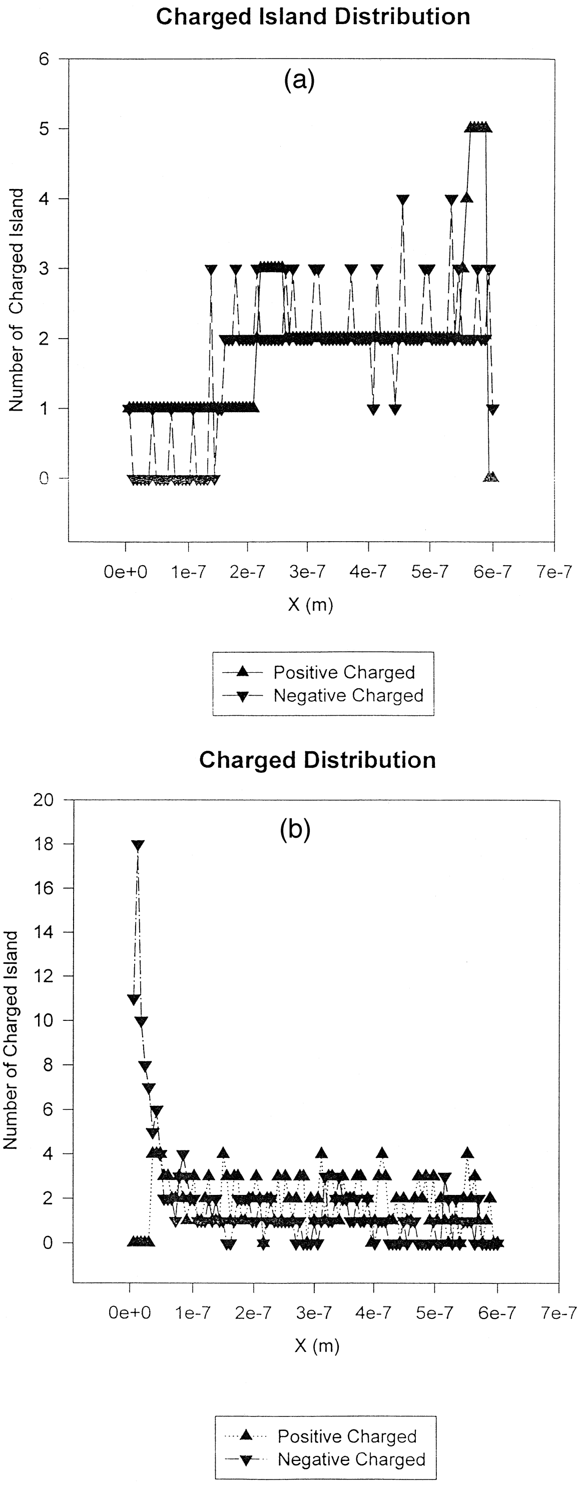

Fig. 4. Charge distributions along film; (number of charged islands in each row across film.) (a) Forward/positive and (b) reverse/negative bias.

across the films is $V = 10^{-4}$ V, eV $\ll kT \ll \delta E$. (Exceptions to these standard parameters are clearly identified in figure captions.)

The number of equilibrium charged islands is calculated, and positive and negative assigned randomly in the array. At $t = 0$, voltage is applied and the initial field distribution determined iteratively for this Boltzmann distribution. Tunnelling probabilities are calculated for all gaps along and across the film, and time is incremented. The probability calculations are repeated at each time increment, and a tunnelling transition occurs whenever the accumulated probability exceeds 0.5. There is a simplifying rule-of-thumb rule: any adjacent positive and negative islands are assumed to immediately recombine, and the underlying Boltzmann distribution re-established by the random assignment of two charged islands elsewhere. Only the tunnelling events in Fig. 1 are considered, i.e. there are no doubly charged islands, and charged/neutral transitions are not activated, ignoring the proposal of a reduced δE due to the difference between static and high frequency values of ε_r [9].

3. Simulation results.

The diode effect (Fig. 2) matches the form of the original, even to the roughly 10:1 conductance ratio. Fig. 3a and b show the potential distributions along the film for the forward and reverse bias directions, respectively. In both cases again, there is a very satisfactory match to the general experimental forms (which is more obvious in a linear format). The effects of charged islands are very apparent at the edges. Positive and negative charge distributions are shown in Fig. 4, corresponding to the data in Fig. 3, and complying with Poisson's Law. It is expected that the systematic study of field and charge distributions at higher applied fields will yield a discontinuity and the reproducible switching effect observed in inhomogeneous films (figure 3b of Ref. [5]). The variation of film conductances with anomalous electrode gap width is shown in Fig. 5. Fig. 6 shows the transient pseudo-inductive effect, with the ratio of final to initial currents. The traditional R–C circuit model for the film would predict quite a different response with an initial transient spike.

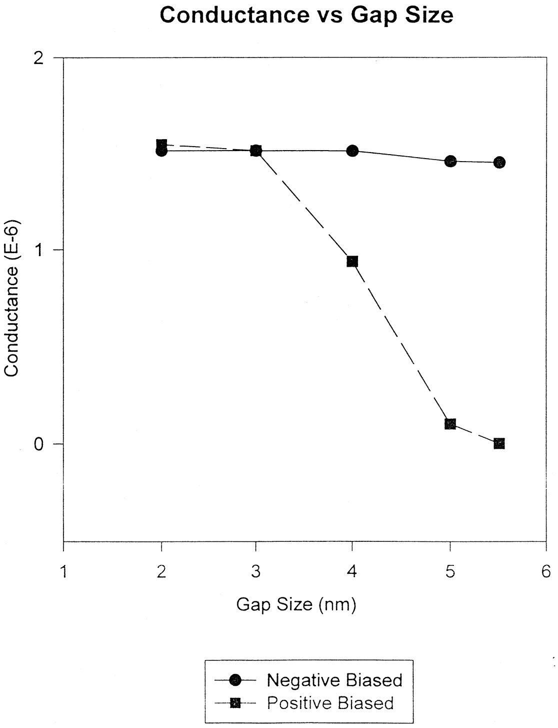

Fig. 5. Conductance variation with electrode gap width to 'anomalous' islands; gap width plus island diameter maintained constant at 6 nm.

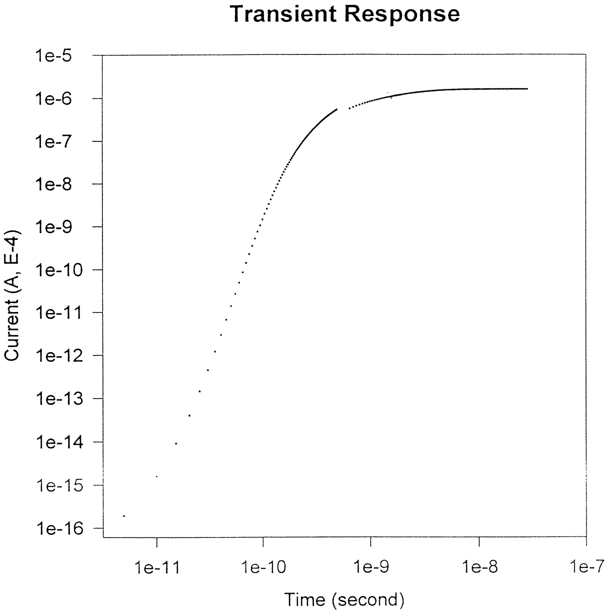

Fig. 6. Transient current response.

4. Discussion

There has been no attempt yet to achieve quantitative agreement with experiment, qualitative matches being considered quite sufficient in the project's initial stages. The purpose of this work is to validate the charge injection model of conduction in discontinuous metal films for sensors [3,4], and the use of island chains and arrays in the single electron tunnelling transistor, where the basic premise is that nanometer scale island tunnelling does not occur until eV exceeds δE, (the 'coulomb blockade') [10]. This condition is not met in this work, which represents a 'sub-threshold' error mechanism.

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