Theory of the current-field relation in silicon-rich silicon dioxide

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The dependence of the current on the electric field in silicon-rich silicon dioxide (Si-rich SiO₂) is studied with the use of a theoretical model based on quantum-mechanical tunneling between a random array of small semiconducting Si islands in a large-band-gap SiO₂ insulator matrix. The current J is calculated in the presence of an electric field F by a simple percolation method for various regimes of external voltage. In the high-field limit, the current is found to obey a Fowler-Nordheim law, $\ln J \sim -\overline{F}/F$, but with \overline{F} weakly dependent on the field F.

I. INTRODUCTION

Mixed phase oxides have recently become of interest in electronic-device applications.¹⁻³ They have been used for field screening in integrated circuits¹ and for charge transfer in electrically alterable read-only-memories (EAROM's).^{2,3} These SiO₂/Si composites have been shown to consist of separate SiO₂ and Si phases.² For $\leq 50\%$ atomic Si in these films, Si islands are typically ≤ 100 Å,² and they are embedded in an SiO₂ matrix. For films chemically vapor deposited (CVD) at 700 °C, the Si islands are amorphous whereas annealing at 1000 °C in N₂ converts many of the islands into crystallites, at least for material containing 45–50 % atomic Si.²

The electrical characteristics of these films have recently been studied (in particular, for films with 34-40% atomic Si), and they have shown a very strong dependence on the average electric field F_{av} for field magnitudes $\geq 1.5 \text{ MV/cm}^4$ Here, $F_{av} = (V_g - \Phi_{ms} - \Psi_s)/l_0$ where V_g is the voltage applied to the gate electrode deposited on top of the off-stoichiometric oxide film, Φ_{ms} is the difference in work function (by convention expressed in volts) between the gate material and the Si substrate onto which the oxide film has been deposited, Ψ_s is the surface potential for the substrate Si-oxide interface, and l_0 is the oxide thickness.⁵ The field dependence of the areal current measured in the external circuit I_{ext} is observed to be exponential in the inverse field; that is, $I_{\text{ext}} \sim e^{-K/F_{\text{av}}}$ where K is \sim constant. These currents are also observed to be only weakly dependent on temperature from 77 K to 300 °C for moderate to large electric field, on contact material for Si, Al, and Au, on voltage polarity, and on oxide thickness in the range from 240 Å to $1 \,\mu$ m.⁴ These experimental results imply that the conduction mechanism in these films is bulk limited by tunneling from Si island to Si island in the off-stoichiometric oxides. Carrierseparation techniques⁶ and electroluminescence⁷ measurements have yielded more evidence to support this conclusion, showing over the moderate field range that carriers are entering the contacts with very little excess energy.⁴ This is what would be expected for dominant islandto-island tunneling as opposed to injection and transport through the oxide phase which can, however, be observed at extremely high average electric fields.⁴ Furthermore,

carrier-separation measurements imply that the dominant carriers transported between the contacts in these films are electrons and not holes, although some hole injection into these films takes place.⁴

In a previous publication, a very simplistic model based on tunneling through a trapezoidal energy barrier was used to relate I to F_{av} , and it was deduced that a $I \sim e^{-K/F_{av}}$ dependence could be obtained with K a weakly dependent function of F_{av} .⁴ In this publication, it was assumed that the local electric field F in the SiO₂ regions between the Si islands would be larger than the average field due to the finite volume of material containing the Si islands and their higher effective dielectric constant, both of which force most of the applied voltage to be dropped across the SiO₂ regions. It is the purpose of this theoretical study to extend this simplistic model over all voltage regions, and to take into account quantum effects arising from Si-island size variations similar to that done by Brodsky for a-Si:H.⁸

II. MODEL CALCULATION

A. Model

The excess silicon in the SiO₂ matrix is formed into spherical islands with a probability distribution P(a) to find an island of radius a. We assume that the distribution is sharply peaked around some average radius a_0 , with a small rms deviation $\Delta a \ll a_0$. The silicon islands are being treated as spherical potential wells, with sharp boundaries. Electrons and holes can be bound by these potential wells (see Fig. 1). The "local" conduction band is the effective potential for electrons, and the local valence band plays the same role for the holes. We denote by U_e and U_h the depths of the potential wells, respectively. Using the standard derivation for the energy levels,⁹ we find for the ground state of the electrons

$$E_{g} \simeq U_{e} \frac{\pi^{2}}{(1+\gamma)^{2}} \left[1 - \frac{1}{3\pi} \left[\frac{\pi}{1+\gamma} \right]^{3} \right],$$

 $\gamma > \pi$
(1)

where, with m_e^* the electron effective mass, and the \hbar Planck's constant,

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FIG. 1. Schematic potentials of a Si island.

$$\gamma = \left[\frac{2m_e^*}{\hbar^2} U_e a^2\right]^{1/2} \tag{2}$$

is the order of 5 for $U_e \sim 3$ eV and $a \sim 10$ Å, and the approximation is good up to 1%. A similar expression can be written for the holes. Above the ground state, we have many bound excited states, which will be approximated as a continuum. Since the depth of the wells is the same i.e., U_e , the ground-state energies (GSE) depend only on the radius a, and thus we get a distribution

$$P(E_g) = P(a) \frac{da}{dE_g} \tag{3}$$

for the energies E_g . This distribution is thus peaked at some E_g^0 , with standard deviation ΔE_g , which can be obtained from P(a).

We assume that the potential wells are randomly distributed in the SiO₂ medium, and the probability of finding a potential well in a volume element dV at a point \vec{r} with GSE between E_g and $E_g + dE_g$ is

$$W(\vec{\mathbf{r}}, E_g) dV dE_g = P(E_g) dV dE_g .$$
⁽⁴⁾

If the average number of excess silicon atoms per unit volume, $N_{\rm Si}$, is given then the average number of islands (potential wells) per unit volume, N_w , can be calculated from

$$N_{\rm Si} = N_w \int dV \int dE_g W(\vec{r}, E_g) = N_w \frac{4\pi}{3} a_0^3 n_0 , \qquad (5)$$

where n_0 is the density of silicon atoms in the island. n_0 is of the order of silicon atomic density in a pure silicon sample; i.e., $n_0 \sim 5 \times 10^{22}$ cm³.

B. Current density

We consider a slab of nonstoichiometric oxide with electrons injected into it, so that an electric field \vec{F} perpendicular to the slab surface is present. The dimensions of the slab are assumed to be large enough so that we can consider the conductivity as a bulk problem. In the presence of the applied field, the injected electrons tunnel from one island to another. Since the current is very small, and thus the number of electrons per unit volume is small, the electrons hop individually from one potential well to another. Most of the time, the potential wells are empty, and we can neglect the correlation between the electrons. We further assume that the presence of an electron in an island causes very small polarization of the medium, and thus the "charging energy"¹⁰ is very small. The reason is that the Si islands in SiO₂ differ considerably from the metallic grains in granular metal films of Ref. 10. While the latter are charged electronic Fermiliquid drops, the Si islands more closely resemble empty potential wells in the SiO₂ matrix. We discuss here only the conductivity associated with electrons.

Since the current consists of hopping between two wells, we first consider two islands of silicon at positions \vec{r}_1 and \vec{r}_2 with radii a_1 and a_2 and GSE E_{g_1} and E_{g_2} in the presence of a strong electric field \vec{F} . The probability of tunneling from the potential well at \vec{r}_1 to the one at \vec{r}_2 is written as (see Ref. 11)

$$T_{12} = Ae^{-(g_{12}^a + g_{12}^o)} . (6)$$

The exponent g_{12}^a is given in the WKB approximation as

$$g_{12}^{a} = 2 \left[\frac{2m_{e}^{*}}{\hbar^{2}} \right]^{1/2} \int_{s_{1}}^{s_{2}} ds [V(s) - E_{g_{1}}]^{1/2} , \qquad (7)$$

where s is integrated along the straight line connecting the two potential wells, s_1 and s_2 are the two ends of this line, and V(s) is the effective potential for the electron along the line. In our simple model, we find with $\Delta V = qFs \cos\vartheta < (U_e - E_{g_1})$,

$$g_{12}^{a} = 2 \left[\frac{2m_{e}^{*}}{\hbar^{2}} \right]^{1/2} \left[(U_{e} - E_{g_{1}})^{3/2} - (U_{e} - E_{g_{1}} - qFs\cos\vartheta)^{3/2} \right] / \frac{3}{2}qF\cos\vartheta , \qquad (8)$$

where s is the shortest distance between the walls of the two islands, ϑ is the angle between the field \vec{F} and the vector $\vec{r}_2 - \vec{r}_1$, and q is the electronic charge.

The second term in the tunneling probability exponen-

tial, g_{12}^b , represents the activation-energy effect. As seen in Fig. 2, it depends on the potential difference (in the presence of the field) between the ground-energy states of the neighboring islands and the temperature T. We ap-



FIG. 2. Schematic potentials and positions of two Si islands.

proximate it as a thermal activation factor, i.e.,

$$g_{12}^{b} = (E_{g_{1}} - E_{g_{1}} - qFs\cos\vartheta)/kT$$
 (9a)

if $E_{g_2} > (E_{g_1} + qFs\cos\vartheta)$, i.e., when the electron has to overcome a positive energy difference by thermal activation, and

$$g_{12}^{b} = 0$$
 (9b)

when $E_{g_2} < (E_{g_1} + qFs \cos\vartheta)$, i.e., when the second island ground-state energy is favorable along the field gradient. In Eq. (9a), k is the Boltzmann constant. Notice that all states above the GSE E_{g_2} of the second potential well are available, but there are no states below it. We have also tacitly assumed that after an electron has tunneled to a potential well it will fall into the local ground state on a time scale much faster than the "tunneling time." Actually, the electron is reaching thermal equilibrium with its new environment, but for simplicity we neglect this and put the electron in the local ground state. This thermal dependence can be considered separately.

The preexponential A is more weakly dependent on the parameters of the problem, and we approximate it as

$$A = E_g / \hbar ,$$

i.e., the average inverse time an electron spends in a potential well.

We now turn to express the total bulk current in the medium. Consider inside the sample an imaginary plane of unit area perpendicular to the field (see Fig. 3). The current density J is equal to the sum of electronic charges which cross the plane per unit time, i.e.,

$$J = q \sum_{i} \sum_{j} T_{ij} , \qquad (10)$$

where *i* is summed over all potential wells on the lefthand side of the unit plane and *j* over wells on the righthand side provided that the line s_{ij} that connects the two wells crosses the plane. In our system of randomly distributed islands, the current density can be cast into the form

$$J = qN_w l \frac{E_g}{\hbar} \sum_j e^{-(g_{ij}^a + g_{ij}^b)}, \qquad (11)$$



CONTROL PLANE



where l is an average typical length, and the j summation is now over the proper right-hand side "neighbors" of the *i*th island on the left-hand side.

C. Percolation treatment

Since the relevant dependence of the current on the parameters of the system is in terms of the exponentials, we find it convenient to relate current flow to the percolation studies of disordered systems.^{11,12} In particular, we should compare our calculation with the results of high-field conduction in disordered systems.^{13,14} We present here a simplified version of the high-field hopping current calculation following arguments similar to Ambegaokar, Halperin, and Langer¹¹ (AHL) and Shklovskii.¹³ In our case, the field affects the tunneling probability both through the effective potential in g_{12}^a and the activation in g_{12}^b .

The essence of the percolation approach is to determine how a set of potential wells, randomly distributed in space, and with random energy distribution, is interconnected by currents flowing from one another. From Eq. (11), we see that the current linking two islands i and j is proportional to

$$T_{ii} \sim e^{-(g_{ij}^a + g_{ij}^b)}$$

and thus varies over many orders of magnitude, depending on the relative positions and energies of this pair of sites. The total current is then obtained by a *summation* of many terms, T_{ij} , which are extremely different in magnitude. It is clear that the "weak" links, i.e., the small current connections are unimportant in this summation, and may be neglected. Effectively only current links with a "strength" above some critical size, $T_{ij} > T_c$, are relevant for the total current; that is, only the islands *i* and *j* with $g = g_{ij}^a + g_{ij}^b$ less than a critical magnitude ζ , play a role in the current flow. The basic assertion of the percolation treatment is that the effective current link T_{ij} can be replaced by $T_c \sim e^{-\zeta}$, since the links with $g \ll \zeta$ are extremely rare, and those with $g \gg \zeta$ are not effective. The essential proposition of the percolation method is that the value of the critical percolation exponent, ζ , can be estimated by geometrical arguments as will follow. Using the percolation prescription, we can now write the current, Eq. (11), as

$$J = q N_w l^* \frac{E_g}{\hbar} e^{-\zeta} , \qquad (12)$$

where all numerical factors of the preexponential are lumped into a "typical length" l^* , which is of order of the interwell distance. Thus we are left with the task of estimating the critical exponent ζ .

The outline of the procedure is as follows: The first step is to calculate the average number of neighbors connected to the *i*th well, so that their T_{ij} is larger than $e^{-\zeta}$, without yet knowing ζ . For a specified model, when the g_{ij} 's of Eq. (11) are given in terms of the positions and energies of the sites, and the probability distribution $W(\vec{r}, E_g)$ of the potential wells is prescribed, the number of "proper" links is

$$n(E_{g_i}) = N_w \int d^3 r_j \int dE_{g_j} W(\vec{r}_j - \vec{r}_i, E_{g_j} - E_{g_i})$$
$$\times \Theta(\zeta - g_{ij}^a - g_{ij}^b)$$
(13)

averaged over the ground-state energies E_{g_i} . In Eq. (13), $\Theta(x)=0$ for x < 0 and $\Theta(x)=1$ for x > 0. The second step is to appeal to a percolation estimate (by geometrical arguments) of the number of relevant neighbors. This number v_c was found to be in the range of 3-5.

In order to calculate $n(E_{g_i})$, we can use g^a of Eq. (8), g^b of Eq. (9), and W of Eq. (4). However, the integration is very involved and tedious, so we perform it approximately in the spirit of AHL.¹¹ First, we will disregard the effect of thermal activation, i.e., concentrate on high fields. Then we will estimate the effect of the activation component g_{ij}^b . Ignoring g_{ij}^b , we find the critical distance s_c so that

Ignoring g_{ij}^{o} , we find the critical distance s_c so that $g_{ij}^{a} < \zeta$. We note that, effectively, only jumps forward (against the field direction) should count, since the high field almost blocks the backward motion. Only angles $\vartheta < \pi/2$ are of interest, and actually small angles are more favorable; therefore, we can avoid the ϑ integration, and set $\cos \vartheta \sim 1$. Now to find s_c , we write

$$2\left[\frac{2m_e^*}{\hbar^2}\right]^{1/2} \frac{\left[(U_e - E_{g_1})^{3/2} - (U_e - E_{g_1} - qFs_c)^{3/2}\right]}{\frac{3}{2}qF} = \zeta ,$$
(14)

where s_c is assumed to be less than $(U_e - E_{g_1})/qF$, i.e., the field F is not strong enough to carry the tunneling electrons into the conduction band of the SiO₂ which separates the islands. In a very small field, where $qFs_c \ll U_e - E_{g_1}$, we find

$$2s_{c} \left[\frac{2m_{e}^{*}(U_{e} - E_{g_{1}})}{\hbar^{2}} \right]^{1/2} = \zeta , \qquad (15)$$

which yields the AHL result $s_c = R_{\text{max}} = \frac{\zeta}{2\alpha}$ where

$$\alpha = \left[\frac{2m_e^*}{\hbar^2}(U_e - E_{g_1})\right]^{1/2} \tag{16}$$

is the inverse length of the tunneling. In the general case, with

$$\eta = qF/(U_e - E_{g_1}) , \qquad (17)$$

we find

$$s_c = \frac{1}{\eta} \left[1 - \left[1 - \frac{3}{2} \frac{\eta}{2\alpha} \zeta \right]^{2/3} \right].$$
 (18)

If we now neglect the effect of g^b on the current (i.e., assume that the activation by the thermal bath is negligible), we find from Eq. (13) in the present approximation

$$n(E_{g}) = 4\pi N_{w} \int dE'_{g} P(F'_{g}) \int_{0}^{s_{c}} ds \, s^{2}$$
$$= \frac{4}{3}\pi N_{w} s_{c}^{3} .$$
(19)

In a more exact treatment of this integration, the size of the potential well a would come into play, but this is neglected here. The percolation prescription gives

$$n(E_g) = \frac{4}{3}\pi N_w s_c^3 = v_c \tag{20}$$

or, the critical length

$$s_c = \left(\frac{3\nu_c}{4\pi N_w}\right)^{1/3} \tag{21}$$

and thus from Eq. (18),

$$\xi = \frac{4}{3} \frac{\alpha}{\eta} \left[1 - (1 - \eta s_c)^{3/2} \right].$$
 (22)

The current density of Eq. (12) is given by

$$J = CqN_w s_c \frac{E_g}{\hbar} e^{-\overline{F}/F} , \qquad (23)$$

where C is a numerical factor of order one, and with

$$F_0 = \frac{4}{3} \left[\frac{2m_e^*}{q^2 \hbar^2} \right]^{1/2} (U_e - E_g)^{3/2}$$
(24)

the Fowler-Nordheim (FN) field, we write

$$\bar{F} = F_0 \left[1 - \left[1 - \frac{qs_c F}{U_e - E_g} \right]^{3/2} \right].$$
(25)

At high fields, \overline{F} depends weakly on F and the current displays a Fowler-Nordheim exponential dependency on the field. However, if the field is moderate, $qs_cF \leq U_e - E_g^0$, the current should deviate from the FN expression. We notice that the effect of percolation, in the

present approximation, is only to replace the average interwell distance

$$s_0 = \left(\frac{3}{4\pi N_w}\right)^{1/3}$$

by $s_c = s_0 (v_c)^{1/3}$. Since this factor appears in the exponential it may be significant. We wish to remark that, since experimentally only the average field F_{av} is measured, while the local field F appears in Eq. (25), it would be difficult to differentiate between the high-field FN regime and the moderate-field percolation regime.

We now turn to a more complete treatment of hopping current, and reconsider the effect of the activation component g^b . We shall provide here only a rough estimate of Eq. (13). For simplicity, we take the bandwidth of the ground-state energies to be ΔE_g [or we assume a square GSE distribution, $W(E_g)=1/\Delta E_g$ in the ΔE_g band and W=0 otherwise]. From Fig. 2, we see that if E_{g_2} is less than $E_{g_1}+qFs\cos\vartheta$, then $g^b=0$, otherwise the tunneling must be activated thermally, g^b is positive, and $n(E_g)$ of Eq. (13) is reduced. We wish to estimate here the reduction factor. If we concentrate on "forward" tunneling (backward tunneling is not effective), we can replace $\cos\vartheta$ by 1, and estimate the average potential drop between two sites by qFs_c , where again s_c is the new critical distance. Thus, a reduction of $n(E_g)$ would occur only when $E_{g_2} > E_{g_1} + qFs_c$. From the Θ function of Eq. (13), E_{g_2} is limited by the condition that $0 \le g^b < \zeta$; i.e.,

$$0 \le \frac{E_{g_2} - E_{g_1} - qFs_c}{kT} < \zeta , \qquad (26)$$

and from bandwidth arguments $E_{g_2} - E_{g_1} < \Delta E_g$. Now we can distinguish two cases: (i) If $(kT\zeta + qFs_c) > \Delta E_g$, the activation factor does not play any role, and we return to the previous case of Eq. (19). (ii) However, if $kT\zeta + qFs_c < \Delta E_g$, the thermal activation would reduce the E_{g_1} integration of Eq. (13) by a factor of the order of

$$\frac{kT\zeta + qFs_c}{\Delta E_g} \tag{27}$$

and the percolation prescription is now

$$\frac{4\pi}{3}N_{w}s_{c}^{3}\frac{(kT\zeta+qFs_{c})}{\Delta E_{g}}=v_{c}.$$
(28)

This equation with Eq. (18) can now be solved for ζ and the percolation current is expressed by Eq. (12).

We consider a few limiting cases of Eq. (28):

(a) A small field at low temperatures. From Eq. (18) for $qFs_c \ll (U_e - E_g)$, $s_c \rightarrow (1/2\alpha)\zeta$; and if further $qFs_c \ll kT\zeta$, we find

$$\frac{4\pi}{3}N_w kT \frac{1}{\Delta E_g} \frac{1}{(2\alpha)^3} \zeta^4 = v_c$$
 (29)

or, with

$$T_0 = \frac{3v_c}{4\pi N_w} \frac{\Delta E_g}{k} (2\alpha)^3$$

of AHL, we find the famous Mott result

$$\zeta = \left(\frac{T_0}{T}\right)^{1/4}.$$
(30)

(b) Stronger fields at low temperatures. Here $qFs_c \ll (U_e - E_g)$, but not so small compared to $kT\zeta$. To first order in $qFs_c/kT\zeta$, we find

$$\zeta = \left[\frac{T_0}{T}\right]^{1/4} \left[1 - \frac{qF}{8\alpha kT}\right], \qquad (31)$$

which is similar to the Pollak and Riess¹⁴ result.

(c) Moderate fields at low temperatures. Here $qFs_c \ll (U_e - E_g)$ and also $kT\zeta \ll qFs_c$, and we find

$$\frac{4\pi}{3}N_w qF \frac{1}{\Delta E_g} s_c^4 = v_c \tag{32}$$

and with $s_c = \zeta/2\alpha$, we find similar to Pollak and Riess,¹⁴

$$\zeta = \left(\frac{F_{\rm PR}}{F}\right)^{1/2}$$

where

$$F_{\rm PR} = \frac{3\nu_c}{4\pi N_w} (2\alpha)^4 \frac{\Delta E_g}{q} . \tag{33}$$

(d) If the field is higher and qFs_c is not much smaller than $U_e - E_g$ but still $qFs_c < \Delta E_g$, we use Eq. (28) for s_c to obtain

$$s_c^4 = \frac{3\nu_c}{4\pi N_w} \frac{\Delta E_g}{qF} \tag{34}$$

and ζ is found from Eq. (22). This yields again a FN dependence of the current as in Eq. (24), but instead of Eq. (25) we have a stronger dependence of \overline{F} on F, i.e.,

$$\bar{F} = F_0 [1 - (1 - \beta F^{3/4})^{3/2}], \qquad (35)$$

where

$$\beta = \frac{q}{U_e - E_g} \left[\frac{\Delta E_g}{q} \frac{3\nu_c}{4\pi N_w} \right]^{1/4}.$$
 (36)

(e) At still-higher fields when $qFs_c > \Delta E_g$, we return to Eq. (25).

III. CONCLUSIONS

A theoretical model based on quantum-mechanical tunneling between an array of small semiconducting Si islands in a large-band-gap SiO₂ insulator matrix to predict current-field dependence over a wide range of applied voltage conditions has been developed. The model was broken up into three distinct voltage regions using a percolation treatment in the low-field range, and the WKB approximation in the moderate-to-high electric field range. The form of the derived *J-F* results in the moderate-to-high field regime and the predicted temperature dependence is in agreement with the observed experimental results for Si-rich SiO₂ films with 34–40 % atomic Si.⁴ No experimental test of the low-field predictions has been performed. Future experiments will be in this direction since the low-field J-F results and their temperature dependence will control the ultimate information

storage ability over long periods of time of certain EAROM's which will use these films.³ This model can be used for any other separate phase semiconductor-insulator system or for cermets (metal-insulator systems).

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