

# Charge transport in dielectrics via tunneling between traps

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A theory for charge transport in dielectrics via tunneling between traps was developed. Unlike in the Frenkel model, in the present theory the traps are characterized with the thermal and optical ionization energies, and ionization proceeds by the multi-phonon mechanism. The theory predicts the tunneling between such traps to be a thermally stimulated process whose activation energy equals half to the difference between the optical and thermal ionization energies. The theory provides an adequate description to the experimental current-voltage characteristics of silicon-rich silicon nitride films. Such films have a high density of traps originating from the excess silicon present in the material, with charge transport proceeding as a charge carriers tunnel between closely spaced traps. © 2011 American Institute of Physics. [doi:10.1063/1.3587452]

## I. INTRODUCTION

Except for thermal oxide  $\text{SiO}_2$  grown on silicon, all other dielectrics, namely, oxides ( $\text{SiO}_x$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Ta}_2\text{O}_5$ ,  $\text{Nb}_2\text{O}_5$ ,  $\text{HfO}_2$ ,  $\text{ZrO}_2$ ,  $\text{TiO}_2$ ) and nitrides ( $\text{Si}_3\text{N}_4$ ,  $\text{SiN}_x$ ,  $\text{Ge}_3\text{N}_4$ , BN) normally have a high density of traps,  $N_t > 10^{19} \text{ cm}^{-3}$ . Today, it is widely accepted that the conduction in dielectrics is controlled by ionization of Coulomb traps according to the Frenkel mechanism.<sup>1,2</sup> The Frenkel effect consists in lowering of the Coulomb barrier for electron emission from a trap in an electric field.<sup>3</sup>

In silicon devices, the two key dielectrics are silicon oxide  $\text{SiO}_x$ ,  $x \leq 2$ , and silicon nitride  $\text{SiN}_x$ ,  $x \leq 3/4$ . In silicon nitride  $\text{SiN}_x$ , injected electrons and holes get readily localized at traps where the trapped charge can be stored for a very long time (about ten years at 85 °C). This effect is used in developing next-generation flash-memory devices.<sup>4</sup> The data storage time in such devices is defined by trap ionization processes. In this connection, the mechanism underlying the transport of electrons and holes in  $\text{SiN}_x$  has recently become a subject of extensive investigation.<sup>5</sup> In spite of numerous studies performed in this field, the nature of traps in  $\text{SiN}_x$  (i.e., their chemical origin) still remains a poorly understood matter.<sup>6,7</sup>

In Ref. 5, an attempt was made to correlate the experimental data for  $\text{SiN}_x$  with the Frenkel model. It was found that, formally, charge transport in  $\text{Si}_3\text{N}_4$  could indeed be described with this model. Yet, for achieving a quantitative comparison between the experimental data and theory for the effective mass of electrons, a value  $m^* \approx (3-5)m_0$  had to be adopted being one order of magnitude greater than the experimental value of this mass,  $m^* \approx 0.5m_0$ . One more difficulty of the Frenkel model consisted in that, for achieving an agreement with the experiment, it was necessary to assume that the attempt-to-escape factor has a physically unreasonable low value of  $\nu = 10^8-10^{10} \text{ sec}^{-1}$ .<sup>5</sup> In the original Frenkel article,<sup>3</sup> it was assumed that the attempt-to-escape

factor has a value of order  $\nu = \phi/h \approx 10^{15} \text{ sec}^{-1}$  (here,  $\phi$  is the trap energy and  $h$  is the Planck constant).

In Ref. 5, we showed that the conduction in  $\text{Si}_3\text{N}_4$  could be quantitatively described with a charge transport theory assuming that the transport process involves repeatedly occurring cycles that comprise electron capture at a trap and subsequent emission of the captured electron from this trap into the conduction band, followed by further capture of this electron at another trap in the downfield region [Fig. 1(a)]. In this theory, a multi-phonon mechanism of trap ionization was assumed. The same trap ionization mechanism was widely used in the interpretation of conduction in semiconductors.<sup>8,9</sup>

It was found, however, that the conductivity of  $\text{SiN}_x$  exponentially increases with excess silicon concentration, or with the parameter  $x$ .<sup>10</sup> So far, this effect has been given no reasonable explanation. The purpose of the present study was to develop a theory for the tunneling transport of electrons between closely spaced phonon-coupled traps [Fig. 1(b)] without intermediate ionization into the conduction band.

The developed theoretical model was then compared with experimental data on the conductivity of  $\text{SiN}_x$  films with various concentrations of excess silicon.

## II. THEORY

A trap will be treated as a single-mode oscillator embedded in the dielectric matrix. We assume that such an oscillator may capture an electron due to a short-range attracting potential. For simplicity, we next assume that this potential is shaped as a three-dimensional  $\delta$ -well in which the bound electron (see Fig. 2) has a single level at energy  $\epsilon$ . Generally, the characteristics of the potential well and, hence, the energy of the bound electron depend on the oscillator coordinate  $q$ . For the sake of convenience, instead of the coordinate  $q$  we will use below a generalized coordinate

$$Q = \sqrt{M}\omega_{ph}q,$$

where  $M$  is the mass and  $\omega_{ph}$  is the oscillation frequency of the oscillator. In this coordinates, the potential energy of a free oscillator has the form

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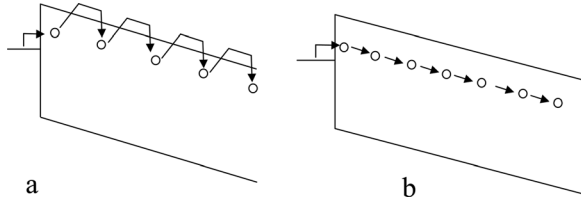


FIG. 1. Electron transport proceeding via electron capture at a trap and subsequent emission of the electron from the trap into the conduction band, followed with subsequent capture of the electron at another trap (a), and electron transport proceeding via tunneling between traps without intermediate emission into the conduction band (b).

$$U_f(Q) = \frac{Q^2}{2}.$$

Usually, the dependence of  $\varepsilon$  on  $Q$  is assumed to be linear, admitting the representation

$$\varepsilon(Q) = -Q_0(Q - Q_0) - W_{opt}.$$

The physical meaning of the quantities  $Q_0$  and  $W_{opt}$  will be clarified below. The potential energy of the oscillator with a bound electron has the form

$$U_b(Q) = \frac{Q^2}{2} + \varepsilon(Q) = \frac{(Q - Q_0)^2}{2} - W_T$$

Thus, the potential energy of an oscillator with a bound electron again has a quadratic form with a minimum,  $-W_T$ , reached at

$$Q_0 = \sqrt{2(W_{opt} - W_T)}. \quad (1)$$

In this position, the electron energy is

$$\varepsilon(Q_0) = -W_{opt}.$$

From here, the meaning of the previously introduced parameters becomes clear: the energy  $W_T$  is the thermal ionization energy of the trap, or the minimum energy required for the electron to escape from the trap, and  $W_{opt}$  is the optical ionization energy (with the optical ionization proceeding via a vertical transition, see Fig. 2). For a  $\delta$ -potential the electron wavefunction, dependent on the radius  $r$  around the trap, has the form

$$\psi(r) = \sqrt{\frac{\kappa}{2\pi}} \frac{e^{-\kappa r}}{r}, \quad \kappa = \frac{\sqrt{-2m * \varepsilon(Q)}}{\hbar}, \quad (2)$$

so that it involves an implicit dependence on the oscillator coordinate

Next, we assume that at a distance  $D$  from the trap a second, empty trap is located. As the oscillator vibrates, the trapped-electron energy  $\varepsilon_1$  changes, so that at some instances the electrons energy becomes coincident with the energy  $\varepsilon_2$  of the empty level due to the second trap:

$$\varepsilon_1[Q_1(t)] = \varepsilon_2[Q_2(t)].$$

At such moments, transitions of trapped electrons from one to another trap become possible. The rate of such transitions is given by<sup>11</sup>

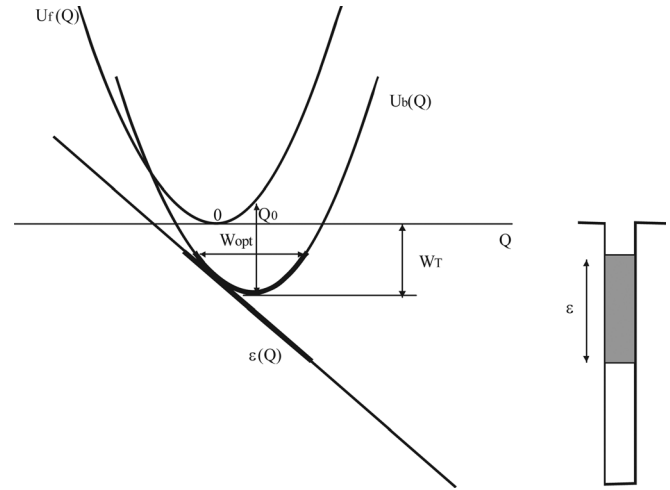


FIG. 2. Configuration diagram of a free oscillator with potential energy  $U_f(Q)$  and that of the same oscillator occupied with a trapped charge, with potential energy  $U_b(Q)$ .  $\varepsilon(Q)$  is the energy position of trapped electron considered as a function of the oscillator coordinate  $Q$ . The thick line shows the correspondence between the range of oscillator coordinate and the range of the electron energy at the trap in the line  $\varepsilon(Q)$ . Shown on the right is a sketch of the potential well for the electron; here, the range of electron level energy  $\varepsilon(Q)$  is shown as a shaded area.

$$\nu = \frac{\pi|V|^2}{\hbar} \delta[\varepsilon_1(Q_1) - \varepsilon_2(Q_2)]. \quad (3)$$

Here,  $V$  is the matrix element of the interaction potential leading to the electron transition from one trap to other. In the case under consideration, this matrix element is the overlap integral of wavefunctions (2) for the electron states localized at the traps<sup>12</sup>

$$V = \frac{\hbar^2 \kappa}{m * D} e^{-\kappa D}. \quad (4)$$

According to the ergodic theorem, for finding the mean rate of the process we have to average expression (3) over the distribution of oscillators in the coordinate-momentum space  $(Q, P)$

$$\bar{\nu} = \int \frac{\pi|V|^2}{\hbar} \delta[\varepsilon_1(Q_1) - \varepsilon_2(Q_2)] f_1(Q_1, P_1) f_2(Q_2, P_2) dQ_1 dP_1 dQ_2 dP_2. \quad (5)$$

Here

$$f_1(Q_1, P_1) = \frac{1}{2\pi kT} \exp \left[ -\frac{(Q_1 - Q_0)^2 + P_1^2}{2kT} \right] \quad (6)$$

is the equilibrium distribution for the first occupied trap (with trapped electron) and

$$f_2(Q_2, P_2) = \frac{1}{2\pi kT} \exp \left( -\frac{Q_2^2 + P_2^2}{2kT} \right) \quad (7)$$

is the distribution for the second empty trap. The distributions (6) and (7) imply that, here, the classical representation for the oscillator motion is used.

The energy of the electron at the second trap depends on the coordinate of the second oscillator

$$\varepsilon_2(Q_2) = -Q_0(Q_2 - Q_0) - W_{opt} - eFD,$$

where the quantity  $eFD$  stands for the drop of the external potential over the trap-to-trap separation ( $F$  is the electric field).

On integration of (5) over  $P_1$ ,  $P_2$ , and  $Q_2$ , we readily obtain:

$$\bar{\nu} = \int \frac{\hbar|\varepsilon|}{m^* D^2 k T Q_0} \exp \left\{ -\frac{(Q - Q_0)^2 + (Q - eFD/Q_0)^2}{2kT} - \frac{4\sqrt{2m^*}}{3\hbar e F} [(-\varepsilon)^{3/2} - (-\varepsilon - eFD)^{3/2}] \right\} dQ$$

$$\varepsilon = -Q_0(Q - Q_0) - W_{opt}. \quad (8)$$

An analysis of the integrand in Eq. (8), easy to perform at zero electric field, shows that the integrand has a sharp maximum at

$$Q^* = \frac{Q_0}{2} \left( 1 - \frac{kT D \sqrt{2m^* W_T}}{W_T \hbar} \right). \quad (9)$$

As a rule, for deep traps hosted in a dielectric the condition  $kT \ll W_T$  is fulfilled and the localization length of a trapped electron is comparable with the separation between traps,  $\lambda = \hbar/\sqrt{2m^* W_T} \sim D$ . It can be inferred from Eq. (9) that, to a good accuracy, tunneling proceeds when both oscillators reach the coordinate  $Q^* \approx Q_0/2$  (see Fig. 3). The integral (8) can be calculated by the steepest descent method. Under the condition  $eFD \ll W_T$ , this calculation yields:

$$\bar{\nu} = \frac{\sqrt{\pi} \hbar W_T}{m^* D^2 Q_0 \sqrt{kT}} \exp \left( -\frac{W_{opt} - W_T}{2kT} \right) \exp \left( -\frac{2D\sqrt{2m^* W_T}}{\hbar} \right) \times \exp \left( -\frac{eFD}{2kT} \right) \quad (10)$$

In the above representation, the tunneling rate acquires quite a clear meaning. The first exponential shows that a certain activation energy,  $(W_{opt} - W_T)/2$ , has to be overcome in the tunneling process. The second exponential is the traditional tunneling factor, the transparency of the tunnel barrier between traps. The third exponential is the barrier lowering in the electric field, with the exponent like that in the Pool law. An estimate of the pre-exponential factor with  $W_T = 1.5$  eV,  $W_{opt} = 3$  eV,  $kT = 0.03$  eV,  $D = 10^{-7}$  cm, and  $m^* = 0.5m_0$  gives  $\nu_0 = 2 \times 10^{15} \text{ sec}^{-1}$ .

In a crude approximation, the current through dielectric varies in the proportion to the rate of electron tunneling between traps. It can therefore be expected that, according to Eq. (10), the current should increase with a trap density due to the decreasing trap-to-trap separation  $D$ , and the current-voltage characteristics should follow the Pool law, i.e., admit linearization in the coordinates  $\lg J - F$ ; in addition, the current should show a growth with an increasing temperature.

In Ref. 5, a theory of charge transport in dielectrics containing phonon-coupled traps was developed in which the transport process was assumed to involve a sequence of trap ionization events followed by a capture of the freed charge at a neighboring trap. The electrons were assumed to be injected from the contact to the conduction band of the dielectric either by the Fowler-Nordheim mechanism or through an intermediate trap.<sup>13</sup> In the present study, we assume that the charge transport inside the dielectric

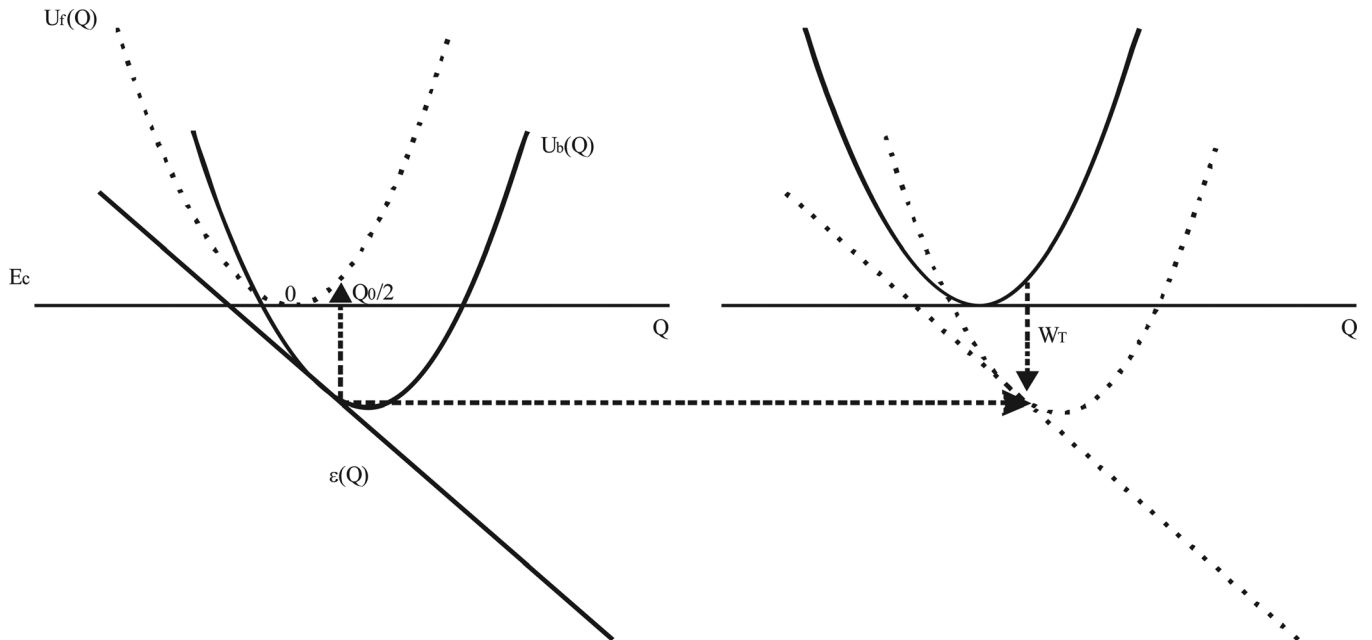


FIG. 3. Configuration diagram for two phonon-coupled traps.  $U_f(Q)$  is the potential energy of an empty oscillator (without trapped electron);  $U_b(Q)$  is the potential energy of an occupied oscillator (with trapped electron);  $\varepsilon(Q)$  is the position of the energy level of trapped electron dependent on the coordinate  $Q$ ; and  $E_c$  is the conduction-band edge. The solid and dotted lines refer to the initially occupied and empty state, respectively. The horizontal dashed arrow shows the most probable tunneling transition for the electron when both oscillators take the position  $Q_0/2$ . The dashed vertical arrows show the oscillator transitions that proceed as electron tunnels from the first to the second trap.

proceeds via the tunneling between traps without electron emission into the conduction band.

In the numerical model for the charge transport we use the following equation for the trap occupation number in the dielectric, assuming that dielectric thickness is much larger than distance between traps:

$$\frac{\partial}{\partial t} n_i = \nu_{i-1,i} n_{i-1} \left(1 - \frac{n_i}{N_t}\right) - \nu_{i,i-1} n_i \left(1 - \frac{n_{i-1}}{N_t}\right) + \nu_{i+1,i} n_{i+1} \left(1 - \frac{n_i}{N_t}\right) - \nu_{i,i+1} n_i \left(1 - \frac{n_{i+1}}{N_t}\right). \quad (11)$$

Here,  $N_t$  is the total density of traps,  $n_i$  is the density of traps occupied with electrons at the  $i$ th node of the calculation domain. The node-to-node separation was chosen to be equal to the mean distance between the traps  $D = N_t^{-1/3}$ . The rates of tunneling transitions from the node  $i-1$  to the node  $i$  and in the reverse direction,  $\nu_{i-1,i}$  and  $\nu_{i,i-1}$ , were calculated by the Eq. (10). The filling of the first trap adjacent to the contact was described with the equation

$$\frac{\partial}{\partial t} n_1 = Inj \times (N_t - n_1) - Ion \times n_1 + \nu_{2,1} n_2 \left(1 - \frac{n_1}{N_t}\right) - \nu_{1,2} n_1 \left(1 - \frac{n_2}{N_t}\right), \quad (12)$$

where the coefficient of trap filling with the electrons from the contact,  $Inj$ , and the coefficient of reverse ionization of the trap into the contact,  $Ion$ , were calculated using the following consideration.

The problem about the rate of ionization of a trap with a single level  $\varepsilon$  from dielectric into the contact was treated in.<sup>12</sup> It was shown that, in a broad range of conditions, this rate could be represented as:

$$Ion(z) = \frac{V_{out}}{2z} \exp \left\{ -\frac{4(2m^*)^{1/2} [(-\varepsilon)^{3/2} - (-eFz - \varepsilon)^{3/2}]}{3eF\hbar} \right\} / \left( 1 + \exp \left( -\frac{\Phi + \varepsilon + eFz}{kT} \right) \right),$$

where  $\Phi$  is the energy difference between the conduction-band bottom in the dielectric and the Fermi level in the contact,  $z$  is the trap-to-contact separation, and  $V_{out}$  is the velocity of free electron in the contact. For a phonon-coupled trap, this rate of ionization needs to be averaged over the distribution function of the oscillator:

$$Ion = \int \frac{V_{out}}{2z\sqrt{2\pi kT}} \frac{\exp \left\{ -\frac{(Q - Q_0)^2}{2kT} - \frac{4(2m^*)^{1/2} [(-\varepsilon)^{3/2} - (-eFz - \varepsilon)^{3/2}]}{3eF\hbar} \right\}}{1 + \exp \left( -\frac{\Phi + \varepsilon + eFz}{kT} \right)} dQ, \\ \varepsilon = -Q_0(Q - Q_0) - W_{opt}.$$

Next, we find the rate of electron injection from the contact to trap  $Inj$  from the condition that, in a steady state, the trap occupation number should obey the statistics governed by the Fermi level in the contact,

$$n_1 = \frac{N_t}{1 + \exp \left( \frac{\Phi - W_T}{kT} \right)}.$$

From this reasoning, using the stationary Eq. (12), without allowance for the transitions to subsequent traps we obtain

$$Inj = e^{-\frac{\Phi - W_T}{kT}} Ion.$$

The distribution of the electric field in the dielectric was found as a result of solving the Poisson equation

### III. EXPERIMENT AND THEORY COMPARISON

The theoretical results were compared to the data obtained in an experiment on measuring the conductivity of

amorphous silicon-rich silicon nitride films deposited in CVD processes held at various  $\text{SiH}_4/\text{NH}_3$  flow ratios.<sup>10</sup> In the experiment, we measured the electric current versus the voltage applied to across the dielectric of thickness  $\approx 100$  nm. It is a well-known fact that an increase in the  $\text{SiH}_4/\text{NH}_3$  ratio during the deposition of  $\text{SiN}_x$  results in a reduction of parameter  $x$ , or in an increase of excess, overstoichiometric silicon concentration. An increase of  $\text{SiH}_4/\text{NH}_3$  ratio is accompanied with increasing of the refraction index  $n$  whose value is indicated in the caption to Fig. 4. Figure 4 shows the current-voltage characteristics measured at room temperature for stoichiometric silicon nitride (1, 2) and for silicon-rich silicon nitride (3, 4).

Engaging attention is the fact that, for silicon-rich silicon nitride, greater current values are observed in comparison with stoichiometric silicon nitride. Moreover, an appreciable electric current across silicon-rich silicon nitride films starts flowing already at low voltages, whereas in the case of stoichiometric silicon nitride, there is a certain voltage threshold for the establishment of a notable current. We

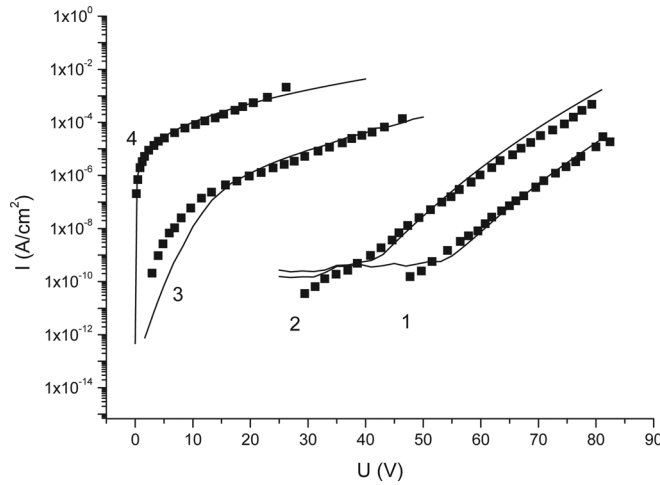


FIG. 4. Experimental (symbols) and calculated (lines) current-voltage characteristics of  $\text{Si}_3\text{N}_4$  with different values of trap concentration. In the calculations, the adopted trap parameters were as follows: 1-  $\text{SiH}_4/\text{NH}_3 = 0.02$ ,  $n = 1.96$ , input parameters:  $N_t = 10^{19} \text{ cm}^{-3}$ ,  $W_T = 1.7 \text{ eV}$ ,  $W_{\text{opt}} = 3.5 \text{ eV}$ ,  $\Phi = 2.3 \text{ eV}$ ,  $m^* = 0.5 m_0$ ; 2 -  $\text{SiH}_4/\text{NH}_3 = 0.1$ ,  $n = 2.05$ ,  $N_t = 2 \times 10^{19} \text{ cm}^{-3}$ ,  $W_T = 1.5 \text{ eV}$ ,  $W_{\text{opt}} = 3 \text{ eV}$ ,  $\Phi = 2.3 \text{ eV}$ ,  $m^* = 0.5 m_0$ ; 3 -  $\text{SiH}_4/\text{NH}_3 = 0.2$ ,  $n = 2.2$ ,  $N_t = 3 \times 10^{21} \text{ cm}^{-3}$ ,  $W_T = 1.55 \text{ eV}$ ,  $W_{\text{opt}} = 3 \text{ eV}$ ,  $\Phi = 2.1 \text{ eV}$ ,  $m^* = 0.5 m_0$ ; 4 -  $\text{SiH}_4/\text{NH}_3 = .5$ ,  $n = 2.35$ ,  $N_t = 3 \times 10^{22} \text{ cm}^{-3}$ ,  $W_T = 1.7 \text{ eV}$ ,  $W_{\text{opt}} = 3 \text{ eV}$ ,  $\Phi = 2.1 \text{ eV}$ ,  $m^* = 0.5 m_0$ . Here,  $\Phi$  is the barrier height for electron injection from silicon substrate into nitride.

attribute such a drastic contrast between the conductivity values of stoichiometric silicon nitride and silicon-rich silicon nitride to the presence of a high density of traps in the latter nitride. This interpretation is consistent with the previously advanced hypothesis<sup>14</sup> that the nature of traps in silicon nitride is related with the presence of silicon clusters in the material. In particular, the smallest silicon cluster, the Si-Si bond, can act as an electron trap in silicon nitride.<sup>15</sup>

We carried out numerical calculations of current-voltage characteristics in a model in which only ionization of traps to the conduction band of the dielectric, i.e., without tunneling between traps, was taken into account. Those calculations showed a good agreement with the experimental data for near-stoichiometric nitride (curves 1 and 2 in Fig. 4); we, however, failed to adequately reproduce the experimental data that were obtained for silicon nitride enriched with silicon. Only the model, taking the tunneling between traps into account, was found capable of providing an adequate description to those data (curves 3 and 4 in Fig. 4). For a good agreement between the theoretical calculations and the experiments it was necessary to assume that a high concentration of traps, in excess of  $10^{21} \text{ cm}^{-3}$ , was present in the material.

#### IV. CONCLUSION

To summarize, it was experimentally and theoretically shown that silicon-rich silicon nitride and stoichiometric nitride are dominated by different conduction mechanisms. Unlike in stoichiometric nitride, where electron transport proceeds via trap ionization through electron emission into conduction band followed by a subsequent capture of the free electron at another trap, in silicon-rich silicon nitride with a high concentration of traps and, hence, with a small trap-to-trap separation the charge transport proceeds as an electrons tunnel from one trap to another without electron emission to the conduction band of silicon nitride. For this process, we have developed a theory giving a description to electron tunneling between phonon-coupled traps. Numerical calculations based on this theory revealed a good agreement of calculated data with experimentally measured current-voltage characteristics of silicon-rich silicon nitride films. The theory predicts the tunneling transitions between phonon-coupled traps to be a thermally stimulated process whose activation energy equals half the difference between the optical and thermal ionization energies of the traps. It should be emphasized here that in the present model, unlike in the Frenkel-Hill model,<sup>1,3</sup> the traps were assumed neutral.

#### ACKNOWLEDGMENTS

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