INVESTIGATION OF ELECTRON AND HOLE TUNNELING THROUGH THIN SILICON DIOXIDE FILMS

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The processes of electrons and holes tunneling through thin (~ 10 nm) silicon dioxide films have been investigated. In parallel with the direct measurement of tunneling current in Si–SiO_2–Al structures, an original method, based on the accumulation of charge in Si–SiO_2–Si_3N_4–Al structures, have been used. The current-voltage dependences obtained by two methods for electron tunneling are in agreement and correspond to carrier injection from silicon at a positive voltage on metal according to the Fowler – Nordheim mechanism. The hole tunneling current can be measured only by the charge accumulation method due to a high energy barrier for holes. Possible mechanisms of positive charge accumulation are discussed. The obtained values of the exponential in the dependence of the accumulation current on reverse field are equal to $7.5 \cdot 10^8$ and $6.8 \cdot 10^8$ V/cm, and are assumed to be due to tunnel injection. In this case, the estimations for the effective masses of holes in the forbidden band of SiO_2 of about $1.2m_0$ and $1.0m_0$ are obtained.

Introduction

The continuous scaling down of MOS (Metal – Oxide – Semiconductor) devices into the deep submicron region requires ultrathin gate dielectrics with high reliability. Thin insulator films of silicon dioxide (SiO_2) are widely used in ULSICs (Ultra Large Scaled Integrated Circuits) as a gate dielectric and in circuits of electrically-alterable read-only memory: MNOS (Metal – Nitride – Oxide – Semiconductor) and floating gate structures.

The mechanism of charge transfer through thin films of thermal SiO_2 is tunnel emission of electrons. In case of ultrathin dielectric films SiO_2, the electron tunneling through a triangular or trapezoidal energy barrier depending on SiO_2 thickness and applied electric field is observed [1 – 4]. It has been theoretically shown that, in case of tunneling through a triangular barrier (Fig. 1), the process is described by the exponential dependence of current on the reverse electric field in a dielectric [5]. Studies have shown that the quasiclassical WKB (Wentzel – Kramers – Brillouin) approximation developed in [5] describes experimental results very well [2, 6 – 8]. The exponent in the Fowler – Nordheim expression for the dependence of current on electric field is determined by the barrier height for tunneling and by the effective mass of carriers in the dielectric. This makes it possible, with a known value of the barrier height, to evaluate the effective mass. For electrons with the barrier height of Si–SiO_2 of 3.1 to 3.25 eV, the effective mass equaled 0.4 to 0.5 m_0, where m_0 is the free electron mass.

The experimental determination of the energy barriers and effective mass of holes from current measurements in MOS structures presents great difficulties. Due to a strong asymmetry of barrier heights $\Phi_h > \Phi_e$, independently of the polarity of the applied voltage, the current of electron injection from a semiconductor or from a metal electrode J_e largely exceeds the current of tunnel emission of holes J_h.

In this work, we have investigated the current-voltage characteristics of electron and hole currents during the tunneling of charge carriers through a triangular barrier of thermal SiO_2 in high electric fields (about 10^7 V/cm). Two types of structures were used: MOS and MNOS. In the MOS structure, the tunnel current was measured by the direct method. In case of the MNOS structure, the injection current was evaluated by a method developed earlier in [7 – 9]. The value of current was found from the quantity of the accumulated charge in the two-layer dielectric (SiO_2-

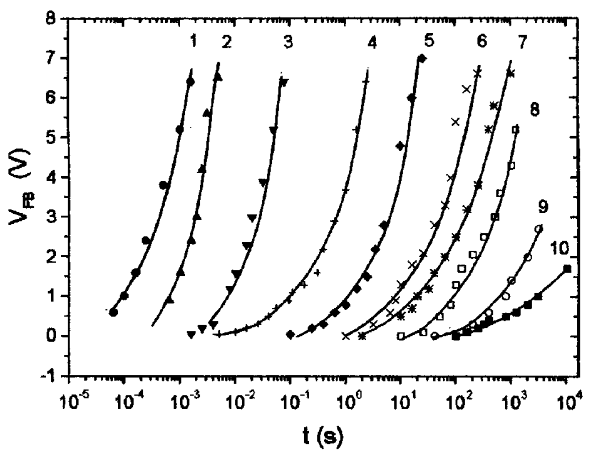

Fig. 1. Accumulation curves of a negative charge at various voltages on the metal electrode (n-type Si(111) d_{ox} = 9.8 nm): V_g = 151 V (*1*), 146 (*2*), 135 (*3*), 124 (*4*), 115 (*5*), 110 (*6*), 107 (*7*), 105 (*8*), 103 (*9*), 100 (*10*)

Si_3N_4) which was determined by a shift of the capacity-voltage characteristic of the structure. The SiO_2 film thickness was about 10 nm. It is much larger than a possible thickness of the transition SiO_x layer at the Si– SiO_2 interface and the oxinitride layer at the SiO_2– Si_3N_4 interface which enable us not to consider them.

1. Experiment

1.1. Samples

The MOS and MNOS structures were prepared on n- and p-type of silicon with doping levels of $1 \cdot 10^{15}$ and $3.5 \cdot 10^{14}$ cm^{-3}, respectively. Wafers after the standard RCA chemical treatment [10] were oxidized in dry oxygen at 900 °C up to the silicon thickness about 10 nm. The SiO_2 thickness was measured using a laser ellipsometer ($\lambda = 632.8$ nm). In case of MNOS structures, the silicon nitride film was grown by the CVD (Chemical Vapor Deposition) method from the $SiCl_4 + NH_3$ reaction at a temperature of 875 °C. The thickness of the Si_3N_4 layer was 300 nm. Such a thick Si_3N_4 film allows one to weak significantly the contribution of a negative charge injected from metal to a shift of the capacity-voltage characteristic during investigation of the hole injection from semiconductor. The area of aluminum contacts was in a range from $0.6 \cdot 10^{-3}$ to $5 \cdot 10^{-3}$ cm^2.

He–Ne laser illumination was used during charge storage when the reverse polarity of electric voltage was applied to exclude a non-equilibrium depletion in semiconductor.

1.2 . Evaluation of Tunnel Injection Current

M O S s t r u c t u r e. The current-valtage characteristics of MOS structures have been measured using a Hewlett Packard 4145 A Analyzer. The voltage with polarity corresponded to the surface accumulation of major carriers applied to the MOS capacitor was increased by 0.05 V steps with a time between steps of 300 mS. To determine the built-in charge in dielectric, the high frequency (1 MHz) capacitance-voltage characteristics were used.

M N O S s t r u c t u r e. The charge per area unit Q accumulated in dielectric, when a pulse of voltage is supplied to the structure, was evaluated from a shift ΔV_{fb} of the high-frequency (1 MHz) current-voltage characteristics from the initial value (Fig. 1). In general

$$\Delta V_{fb} = Q / C_m \, (1 - X'_n / d'_n), \tag{1}$$

where $d'_n = d_{ox}\,\varepsilon_n / \varepsilon_{ox} + d_n$, d_{ox} and d_n are the layer thicknesses of oxide and nitride of silicon, respectively, $\varepsilon_n / \varepsilon_{ox} = 1.8$ is the ratio of their relative dielectric constants, C_m is the maximum sample capacity per unit area, $X' = d_{ox}\,\varepsilon_n / \varepsilon_{ox} + X$, X is the distance from the $SiO_2 - Si_3N_4$ interface to the centroid of a trapped charge. The current of charge accumulation on traps is

$$J_{ac} = dQ / dt. \tag{2}$$

It will be assumed that the carriers injected from semiconductor are completely trapped in silicon oxide or nitride near the $SiO_2 - Si_3N_4$ interface. This condition is well fulfilled for structures with a tunnel-thin SiO_2 sublayer with injected charge up to 10^{-6} C/ cm^2 [11]. In this case, it may be assumed that the injection current is $J_{e(h)} = J_{ac}$.

Since structures with relatively thick silicon nitride films were used for measurements, it may be assumed with good accuracy that $X'/d_n \ll 1$ and then

$$Q \approx C_m \, \Delta V_{fb}. \tag{3}$$

The dependence of the shift ΔV_{fb} on $\lg(t)$ was obtained in experiment, t is the resulting duration of the voltage applied to the structure. Then Eq. (2) takes the following form:

$$J_{e(h)} = C_m \, r(E) / (2.3\, t(E)), \tag{4}$$

where $r \equiv \partial(\Delta V_{fb}) / \partial(\lg t)$ is the rate of charge accumulation, E is the electric field in dielectric.

Furthermore, the tunnel current density can be received at the initial stage of charge storage, when the time dependence of ΔV_{fb} may be considered to be linear. In this case, we have

$$J_{e(h)} = \partial(C_m \, \Delta V_{fb}) / \partial(t) = C_m \, r'(E) \tag{5}$$

where $r' \equiv \partial(\Delta V_{fb}) / \partial(t)$ is the slope of the linear part of the $\Delta V_{fb}(t)$ curve. The tunnel current densities obtained with two approaches described were found to be equal.

Relation (2) allows the determination of experimental values of the injection current and is not connected with any mechanism of charge transfer.

The obtained experimental data will be compared with the simple Fowler – Nordheim theory, according to which the injection current through a triangular barrier is

$$J = J_0 \exp(- E_{FN} / F_{ox}), \tag{6}$$

where

$$E_{FN} = 4/3\,(2m^*)^{1/2}\,\Phi^{3/2}/\,\hbar q \quad (7)$$

and E_{ox} is the electric field in the oxide. Here, m^* is the effective mass of carriers in the forbidden band of dielectric, q — the electron charge, Φ — the energy barrier height for tunneling, $\hbar = h/2\pi$ — the Dirac's constant, h — the Planck's constant. Here and further, we omit the indices e and h in the quantities J, m^*, and Φ when speaking about both electrons and holes. For the barrier height $\Phi = 3.25$ eV and $m^* = 0.42\,m_0$, $E_{FN} = 2.6 \cdot 10^8$ V/cm. The pre-exponential factor J_0 depends on the electric field: $J_0 \sim E^2_{ox}$.

2. Results and Discussion

2.1. Electron Tunneling

M O S s t r u c t u r e. For the energy barrier height determination, the current-voltage characteristics of MOS structures were presented as Fowler-Nordheim $\lg(J/E^2) - 1/E$ plots. The slope of Fowler — Nordheim curves allows us to determine the energy barrier heights at the Si—SiO$_2$ or SiO$_2$ — metal interfaces. But, in this case, the value of electron effective mass must be known.

The experimental results for the electron tunneling through thin (10 nm) oxide layers are presented in Fig. 2 (curves *3, 4*). Energy barrier height for electrons at the Si—SiO$_2$ interface estimated from the slope of Fowler — Nordheim curves is equal to 3.2 eV, that agrees with the literature data [6].

During the determination of the energy barriers for electrons on the Si—SiO$_2$ interface, the influence of a fixed charge Q_f in oxide measured with capacitance-voltage characteristics and difference of silicon and metal work function were taken into account. In the case where charge is built-in in oxide with the areal density Q_f and centroid position x_b as referred to the cathode interface, the effective oxide electric field $E_{ox} = V_{ox}(d_{ox})/(d_{ox})$ is no longer equal to the cathode electric field:

$$E'_{ox} = E_{ox} - Q_f/\varepsilon_{ox}\,(1 - x_b/d_{ox}) \quad (8)$$

where ε_{ox} is the oxide permittivity. The position of the fixed charge centroid at half of the oxide thickness has been assumed.

Taking into account φ_{ms} gives

$$E_{ox} = (V_g - \varphi_{ms})/d_{ox}. \quad (9)$$

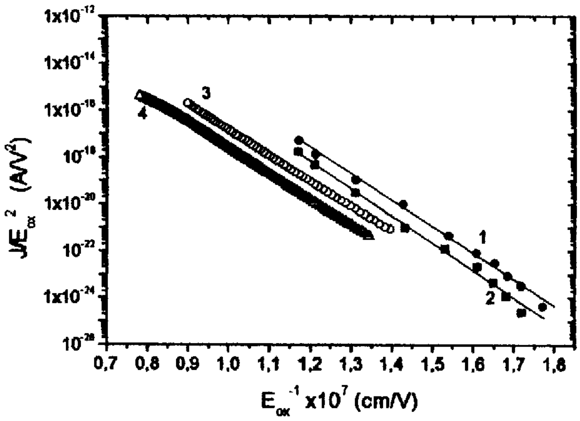

Fig. 2. Dependence of the electron injection current from silicon on the reciprocal electric field in oxide. *1, 2* — data obtained from Fig. 1 according to Eqs. (1) — (4) (*1* — $\Delta V_{fb} = 1$ V, *2* — $\Delta V_{fb} = 3$ V); *3, 4* — direct measurements of electron currents in MOS structures (*3* — *n*-Si—SiO$_2$—Al structure, *4* — *p*-Si—SiO$_2$—Al structure)

M N O S s t r u c t u r e. To see that the electrophysical properties of a 10 nm oxide on which silicon nitride is deposited coincide with the properties of thick SiO$_2$ films, one should first examine the accumulation of negative charge. Fig. 1 shows the accumulation curves $\Delta V_{fb}(\lg t)$ for fields in oxide of $6 \cdot 10^6$ to $9 \cdot 10^6$ V/cm. The accumulation current was calculated from these curves according to (2) — (4) at levels $\Delta V_{fb} = 1$ V and $\Delta V_{fb} = 3$ V. Its dependence on the reverse electric field is given in Fig. 2 (curves *1, 2*). The slope of the experimental line gives the value of the exponential for electrons, $E_{FNe} = 2.6 \cdot 10^8$ V/cm. As can be seen, the data obtained by our technique for the MNOS structure agree well with the measurement of direct currents on MOS structures.

The quantity E_{FN} may be estimated in another independent way similar to that in [12]. The trapped charge changes the field at the contact and, hence, the injection current,

$$J = dQ/dt = J_0 \exp(-E_{FN}/(E_{ox}(0) - Q(t)/\varepsilon_{ox}\varepsilon_0). \quad (10)$$

Here, $E_{ox}(0) = V_g/d'_{ox}$ and ε_0 is the vacuum permittivity. Then if $Q/\varepsilon_{ox}\varepsilon_0 << E_{ox}(0)$,

$$Q = Jdt \approx \varepsilon_{ox}\varepsilon_0 E^2_{ox}(0)/(E_{FN}\ln(1 + t/\tau)), \quad (11)$$

where τ is a parameter independent of t and Q. Then

$$\Delta V_{fb} = Q/C_m = r_0 \lg(1 + t/\tau), \quad (12)$$

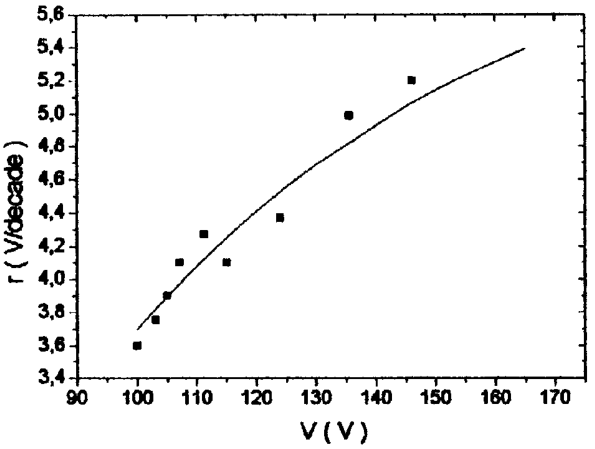

Fig. 3. Negative charge accumulation rate against external applied voltage at a level $\Delta V_{fb} = 3$ V (n-type Si(111) $d_{ox} = 9.8$ nm). The solid line is the calculated dependence $r = r_0 \ (1 - 10^{-\Delta V_{fb}/r_0})$ for $E_{FN} = 2.6 \cdot 10^8$ V/cm and points are the experimental data

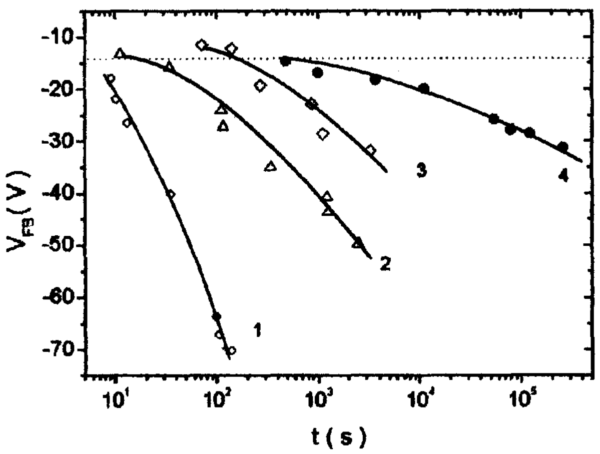

Fig. 4. Positive charge accumulation curves for sample 2 (p-type Si(100)–SiO_2–Al, $d_{ox} = 9.9$ nm). The voltage applied to the structure during the polarization: $V_g = 273$ V (*1*), 255 (*2*), 244 (*3*), 232 (*4*). The dashed line is the level of the initial value of flat-band voltage.

where

$$r_0 = 2.3 \, \varepsilon_{ox} E^2_{ox} (0) \, d'_n / (\varepsilon_n E_{FN}), \qquad (13)$$

the rate of charge accumulation from (12) is

$$r = \partial \Delta V_{fb} (V_g t) / \partial \lg t = r_0 t / (2.3 (\tau + t)) =$$

$$= r_0 (1 - 10^{-\Delta V_{fb}/r_0}). \qquad (14)$$

The dependence $r(r_0)$ at a given level of ΔV_{fb} according to (14) and (13) is determined by the experimental E_{FN}.

Fig. 3 shows the experimental values of the rate of charge accumulation found from Fig. 1 at the level of $\Delta V_{fb} = 3$ V. The dashed curve denotes the dependence $r(V_g)$ calculated according to (14) at $E_{FNe} = 2.6 \cdot 10^8$ V/cm, which coincides with the value of this parameter found from the slope of the dependence $J(1/E_{ox})$ (Fig. 2).

The data given indicate that the injection current from the subsrate is determined by the properties of SiO_2 but not those of silicon nitride.

2.2. Hole Tunneling

M O S s t r u c t u r e. Due to a strong asymmetry of barrier heights $\Phi_h > \Phi_e$, independently of the polarity of the applied voltage, the current of electron injection from a semiconductor of from a metal electrode J_e largely exceeds the current of tunnel emission of holes J_h. In the MOS structure, there is no possibility to measure the hole tunneling current. The hole tunneling into SiO_2 was observed in [12] only when a negative corona charging of the unmetallized surface was used.

M N O S s t r u c t u r e. A set of accumulation curves of positive charge for sample 1 (p-type Si(100) $d_{ox} = 9.9$ nm) is shown in Fig. 4. As can be seen, with a hegative voltage in the metal, a considerable positive charge up to 10^{-6} C/cm may be accumulated in the structure. Fig. 5 shows a similar set of accumulation curves for sample 2 (n-type Si(111) $d_{ox} = 9.8$ nm) at a level of injection lower by an order. It is well seen that, at $t \approx 1$s, a negative charge is accumulated in the structure, and the quantity of this charge is relatively constant. The negative charge accumulation appears to be due to the electron injection current from the metal J_e (Fig. 6,*a*). This injection determines the direct current through the structure.

At times of 1 to 10^4 s depending on field, a positive charge begins to be accumulated. The dependence $\lg J/E^2 - 1/E$ plotted according to (4) from Figs. 4,5 is given in Fig. 7. The value of the exponential determined by the slope of the dependence $\lg J/E^2 - 1/E$ proved to be almost the same for two groups of samples and equal to $6.8 \cdot 10^8$ V/cm (sample 1) and $7.5 \cdot 10^8$ V/cm (sample 2), though the spread in the absolute quantity J for different samples can be about an order.

To analyze the derived dependence $\lg J/E^2 - 1/E$, one should know the localization of the accumulated charge. For this purpose, the discharge of the structure was studied when low voltages of different polarities were applied to the structure [7, 13]. The typical curve of discharge is shown in Fig. 8. At times $\approx 10^3$ s, a 'shelf" appears, the spread of which for different cases is indicated in the figure. The value of the 'shelf" is independent of the quantity of the trapped charge (within the limits of measurements, $\Delta V_{fb} = 30 \div 60$ V). There is no correlation of the 'shelf" depending on the voltage at which the charge accumulation occurs. The discharge curve does not change also in the case where the sample is at the temperature of liquid nitrogen (the charge accumulation occurred at room temperature). This is indicative of the fact that the process of back-discharge occurs according to the tunnel mechanism and cannot be associated with neutralization of a positive charge under the electron drift from the silicon nitride volume to SiO_2, as this current is too small in fields of 10^6 V/cm [13].

When the voltage of $+40$ V was applied to the structure, a sharp acceleration of discharge was observed. The discharge curve was shifted in this case by two orders towards a shorter time. The voltage of negative polarity has a little influence on the discharge. The conclusion should be drawn that the positive charge leaks to the silicon substrate. From the position of the 'shelf", one can roughly estimate the depth of the dielectric layer, at which the tunneling drift of carriers from the traps occurs. With tunneling through a rectangular barrier, the typical time of tunneling will be

$$t \sim \nu^{-1} \exp (d/\lambda), \tag{15}$$

where $\lambda \approx 0.1$ nm, $\nu \approx 10^{13}$ s^{-1} is the frequency factor, d is the barrier thickness. For $t \approx 10^3$ s, we obtain $d \approx 4$ nm. This means that if the positive charge accumulation is due to the tunneling of holes, they are immediately trapped in the SiO_2 film after having passed through the Si–SiO_2 barrier, as the width of a triangular barrier of 4.6 eV in height is also ~ 4 nm for fields of 10^7 V/cm.

Fig. 6 shows the possible mechanisms of accumulation of a positive charge in SiO_2. The possible processes occuring there are considered in [14]. The given mechanism of charge transport should explain, first of all, the magnitude of the slope of current-voltage characteristics *1, 2* given in Fig. 7 as well as the value of the pre-exponential factor. Since the value $E_{FNh} = 7.5 \cdot 10^8$ V/cm obtained for this characteristic is very large, the positive charge accumulation at the expense of ionization of donor traps (current J_t in Fig. 6,*a*) is excluded. Actually, it follows from the obtained value of E_{FNh} that the trap depth should amount to about 7 eV, which is close to the value of the width of the forbidden SiO_2 band. The mechanisms of formation of slow surface states with a strong

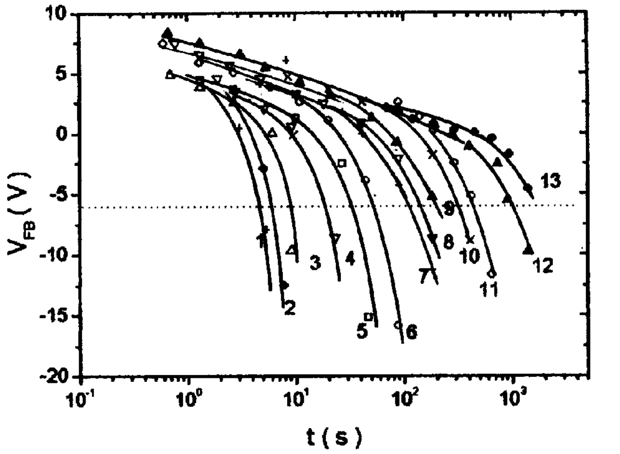

Fig. 5. Positive charge accumulation curves for sample 1 (n-type Si(111)–SiO_2–Al, $d_{ox} = 9.8$ nm). The voltage at the structure during the polarization: $V_g = 270$ V (*1*), 255 (*2*), 250 (*3*), 245 (*4*), 230 (*5*), 290 (*6*), 270 (*7*), 250 (*8*), 230 (*9*), 215 (*10*), 200 (*11*), 180 (*12*), 170 (*13*). The dashed line is the level of the initial value of flat-band voltage

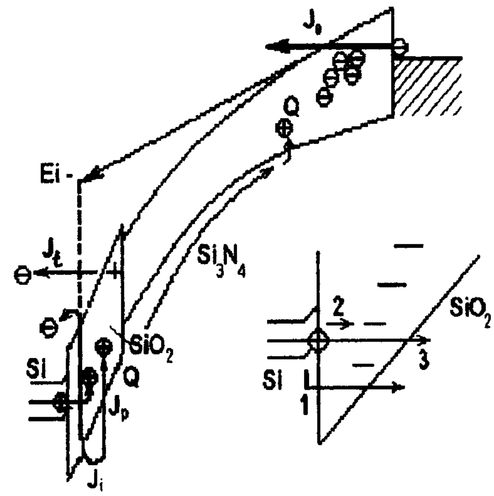

Fig. 6. Possible mechanisms of positive charge accumulation under hole tunneling: *1* – tunneling of hot holes; *2* – direct tunneling into traps; *3* – tunneling into the valence band of SiO_2

exponential dependence on field are also unknown. After the analysis of the mechanism of impact ionization, we obtained the anomalously high value of the exponential in the field dependence of the impact ionization coefficient.

An attempt to obtain the parameters of tunnel transparency of SiO_2 for holes has been made in [12]. The authors used an expression coinciding in essence with (12). The parameters τ_0 and τ found from the experimental dependence $V_{fb}(\lg t)$ were used to obtain

$$J_h = 6 \cdot 10^3 \exp (-3.7 \cdot 10^8 / E_{ox}) \tag{16}$$

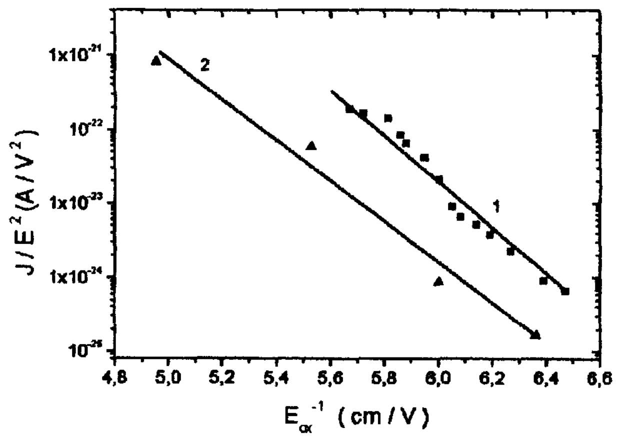

Fig. 7. Dependence of the positive charge accumulation current on the reciprocal electric field in oxide: *1* – sample 1 (*n*-type Si(111)–SiO_2–Al, d_{ox} = 9.8 nm); *2* – sample 2 (*p*-type Si(100)–SiO_2–Al, d_{ox} = 9.9 nm)

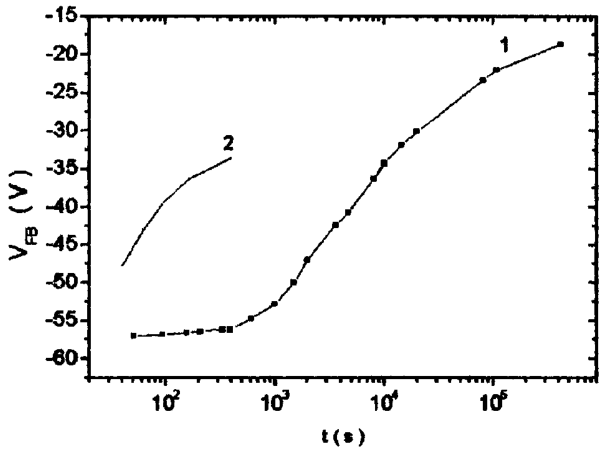

Fig. 8. Discharge curve of the positive charge accumulation (1) and discharge curve (*2*) at the accelerating voltage of + 40 V at Al

(E_{ox} in V/cm, J_h in A/cm^2). However, the calculation of J_0 and E_{FN} only from one curve of accumulation is not very reliable. The rate of accumulation of a positive charge is just close to the theoretical estimate, but there is no such good agreement with the dependence calculated from (14) as for electrons. In fact, it follows from (14) that the maximum value of the accumulation rate is $r_m = r_0 (V_g)$. For our range, r should be 10 to 15 V/decade. In most cases, the value $r \approx 40$ V/decade is achieved. The charge accumulation rate in [12] was 12 V/decade. In our case, an increase of the accumulation rate as compared to the theoretical value is possible due to the fact that the field near the Si_3N_4–Al interface grows with hole accumulation, which results in a decreasing negative charge in the Si_3N_4 volume. This is reflected as a more intensive 'accumulation“ of a positive charge.

From $E_{FNh} = 7.5 \cdot 10^8$ V/cm and $E_{FNh} = 6.8 \cdot 10^8$ V/cm, one can estimate the effective mass of holes in SiO_2. For the barrier $\Phi_h = 4.6$ eV. The obtained value is $1.2m_0$ and $1.0m_0$, correspondingly. The experimentally obtained data on the absolute current quantity (dependences *1*, *2* in Fig. 7) can also be compared to the calculated ones. The comparison shows that the calculated dependence with $m_h = 1.2m_0$ corresponds to these experimental data. However, an estimation of the quantity m_h^* from the absolute value of the injection current cannot pretend to high accuracy, as we do not take into account such effects as a barrier decrease due to the Schottky effect and quantization of levels near the Si–SiO_2 surface. Note that the value $m_h^* = 1.2m_0$ obtained here is also possibly lowered. Electrons, when going from SiO_2 to Si, produce electron-hole pairs. Thus, hot holes can tunnel in SiO_2 (*1* in Fig. 6,*b*), i.e., the real barrier in tunneling can be less than 4.6 eV. Accordingly, the quantity m_h^* will increase. In a similar way, the estimation for m_h^* will change, if holes are trapped in SiO_2 by direct tunneling to traps (*2* in Fig. 6,*b*), not going out to the valence band. In this case, the tunneling goes through a trapezoidal barrier rather than through a triangular barrier.

Conclusion

The electron and hole tunneling through thin dielectric films of SiO_2 has been investigated. The direct measurement of the electron current in MOS structures has been carried out, and the dependences of the current of negative and positive charge accumulation on field with positive (negative) bias on the metal have been obtained in an explicit form from the data on charge accumulation in the dielectric layer of the MNOS structure with a 10 nm SiO_2 sublayer. The results on electron tunneling obtained on MOS and MNOS structures by two different measurement methods are in good agreement. The data on the back-discharge of trapped holes correspond to the assumption that holes are trapped in SiO_2 at a depth of ≈ 4 nm from the Si–SiO_2 interface. The obtained current-voltage characteristics may be interpreted from the standpoint of direct tunneling of holes from silicon to the valence band of SiO_2. In this case, the experimental value of the pre-exponential factor with allowance for a barrier decrease is close to the theoretical value. The values of the effective mass of holes in SiO_2 equal to $1.2m_0$ and $1.0m_0$ follow from the slopes of the dependence $\lg J/E_{ox}^2 (1/E_{ox})$.

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