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I-V CHARACTERISTICS OF THIN FILM Pt/Ba$_x$Sr$_{1-x}$TiO$_3$/Pt STRUCTURES UNDER THE IMPACT OF ELECTRON BEAM IRRADIATION

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A detailed theoretical study of the leakage currents in ferroelectric Pt/Ba$_x$St$_{1-x}$TiO$_3$/Pt (Pt/BSTO/Pt) thin film structures is presented, when the film is irradiated by an electron beam. It is shown, that both the Schottky barrier thermal/field assisted emission and Poole-Frenkel (FP) emission from the oxygen vacancy conditioned electron traps may take place simultaneously if the absorbed energy of electron's from the electron beam is higher than its activation energy to release to the conduction band of the ferroelectric.

Keywords: leakage current, ferroelectric, trapping center, Schottky barrier, Poole-Frenkel emission, oxygen vacancy.

Introduction. The leakage current in thin ferroelectric film devices (memory cell, varactor etc.) has been a subject for extensive studies [1-7]. The magnitude of the leakage current and the shape of the *I-V* curve depend on the conduction mechanism of the dielectric process in the ferroelectric and of the difference in the work function between the electrode and ferroelectric, nature and density of the interface states. Schottky emission [5,8,9], Frenkel–Poole emission [10,11], tunneling (field) emission, space-charge limited regime [1-5, 12], ionic conductance, hopping conductance etc. have been considered. They are characterized by their voltage and temperature dependencies:

$$T^2 \cdot exp\left(\beta V^{\frac{1}{2}}T^{-\frac{1}{2}} - \frac{q\Phi}{kT}\right), V \cdot exp\left(2\beta V^{\frac{1}{2}}T^{-\frac{1}{2}} - \frac{q\Phi}{kT}\right),$$

$$V^2 \cdot exp\left(-\frac{Z}{V}\right), a \cdot \left[\frac{V}{d} + \beta\left(\frac{V}{d}\right)^2\right],$$

where k is the Boltzman's constant, T is the absolute temperature, q is the electron charge. In general, it is not easy to distinguish between these mechanisms. Typically, at lower fields and temperatures (*300 K* and *$10^4 – 10^5$ V/cm)*, the Schottky emission dominates, while at high fields (*10^6 V/cm*) and high field and temperatures, the Frenkel-Poole emission becomes dominant. The Schottky emission and space-charge limited current are not independent. The main experimental results reported for BST and PZT indicated a strong correlation of the leakage current with crystalline structure of the film [1-12]. Currently, it is well established that the oxygen vacancies play a major role. The oxygen concentration is not constant throughout the film. It decreases

sharply near the metal electrodes [1, 4, 7, 12, 13-15], approximately 50% of its value in the center (~20 *nm* from the Pt surface). This oxygen deficient region in the film may have n–type conductivity in contrast to the p–type in the bulk of the film, where the density of the oxygen vacancies is not high. In oxide perovskite ferroelectrics, there are three oxygen ions per unit cell. The density of the oxygen ions in the bulk of the film is $1,5 \cdot 10^{28}$ *m³* [7-9], while in the 20 *nm* thick 50% depleted (by oxygen) interface layer it is $3 \cdot 10^{20}/m^2$. The missing oxygen results in an image charge on Pt electrode. The surface density of the changes on Pt electrode is $3 \cdot 10^{16}$ *oxygen/cm²*. However, not all oxygen vacancies trap electrons. The increased density of the oxygen vacancies at the interface results in a dipole layer-lowering the electrical field in the film (due to the increase of the voltage drop over the Pt/BST interfaces). The non-uniform distribution of the oxygen vacancies near the interfaces causes bending of the energy bands and changes the shape of the barrier, making it for charges easier to overcome. On the other hand, the experimentally observed leakage currents may be explained by conductivity associated with the oxygen vacancies [1-4,13-18]. The oxygen vacancies are the most mobile in perovskite ferroelectrics. The experimental results of both monocrystalline and polycrystalline BSTO at high temperatures [1-4, 7-9] shows that the conductivity of undoped polycrystalline titanate oxides depend on oxygen partial pressure of the ambient gas. At sufficiently high temperatures, the oxygen vacancies are double ionized, each supplying two electrons to the conduction band. This process at low oxygen partial pressure can be described as [4, 7-9]: $O_2 \rightarrow 0.5 O_2 + V^{++}_o + 2e^-$. Oxygen vacancies act as donors [1-6, 13-16] causing n – type conductivity. Moreover, depending on the density of the oxygen vacancies and the density of the background impurities, the conductivity may change from n– to p-type [7, 8, 16, 17]. The interfacial vacancies cause distortion of the crystal lattice and polarization fields around the vacancy. This makes the levels deeper and causes them to act as charge traps [16, 17]. The interfacial built-in electric fields associated with the trapping centers and oxygen vacancies results in [16] changes in the interfacial permittivity of the films [18]. It is worthwhile to notice that the oxygen vacancies are not to be only the main defect of ferroelectric films. The Ba, Sr vacancies in BSTO, result in the shallow acceptor levels. Dopants also result in levels in the forbidden band. Nb gives a shallow level [1, 16, 19], while Mn, Gr, and Fe give levels near midgap. Pt gives a deep level near midgap, too [1, 16, 19].

Most of the metal-ferroelectric junctions lie between the Schottky and Bardeen limits. In Pt – $SrTiO_3$ contact, the effect of vacancies on the barrier height becomes important for the density of vacancies $10^{18}–10^{22}$ *cm⁻³* [1-9, 19]. The, electrochemical interaction of the electrodes with carriers in the ferroelectric may induce "dead layers" [19, 20]. In the case of interfacial surface states and/or dopants there may be a transfer

of charge, and the barrier height cannot be defined as a difference between the metal work function Φ_m and the insulator electron affinity χ [19, 20]. In this case, the resulting barrier height is: $\Phi \cong S(\Phi_m - \Phi_S) + (\Phi_S - \chi) + S\Phi_S$. The calculated barrier height for $SrTiO_3$ on P_t is about 0.9 eV [1, 7], which is close to the 0.8 eV found by photoemission and the 1.1 eV [1-5, 19, 20]. In summary, for low density of oxygen vacancies[1], the undoped ferroelectrics titanates films are considered to have slightly p–type conductivity due to the background impurities (i.e. [Na^+ for Pb^{+2}, Fe^{+3} for Ti^{+4} [1,7,16,19,20]). In reality, most of the ferroelectric films are reach in oxygen vacancies, especially at the interfacial with the electrodes regions. The oxygen vacancies act as donor ions, causing n–type conductivity. In fact, the Kelvin probe study [1, 7, 12, 19, 20] proved that in Pt -$BaTiO_3$ thin film structure, the work function changes from 2.5± 0.3 eV for surfaces to 4.4± 0.4 eV the bulk of the material.

1. The proposed model of the thin film Pt/Ba$_x$St$_{1-x}$TiO$_3$/Pt structure. The three layer model to be considered consists of metal electrodes, Fig.1, n-type interfacial oxygen vacancy rich layers (uncompensated donor concentration n_d with the thickness of δ_l, with the homogeneous distribution of the vacancies, and a poor p–type ferroelectric film (core) [21]. It is assumed, that the concentration of the oxygen vacancies is large at the interfacial with the metal (Pt) contact region, and that some of these vacancies trap electrons and create space–charge regions.

For simplicity, both metal/ferroelectric contacts assumed to be identical. It is also assumed that there are interfacial surface states between the metal contacts and n-type interfacial regions. The thickness of these layers with the surface states is denoted by δ_0. The density of the oxygen vacancies in the middle part of the ferroelectric film (core) is assumed to be relatively low. The work function ($\sim$ 5.6 eV) of Pt is greater, than that of the ferroelectric film ($\sim$3.2 eV), which means that the currents will be emission limited. In equilibrium and at low temperatures all of the trapped electrons would be in traps: at high temperatures and in the presence of an applied field and electron beam irradiation most of these electrons will be excited into shallow traps to the conduction band, either due to the action of the field and electron beam interaction.

The *I–V* characteristic of the proposed model is considered neglecting the diffusion currents associated with the charge concentration gradients. It is also assumed that electrons flow generally by Poole–Frenkel emission (Schottky intrinsic emission) under the influence of simultaneously applied electric field and electron beam irradiation. Additionally, it is assumed that electrons are released from the trap due to absorption of enough energy from the electron beam and can move in the electric field so that the re-trapping process may be neglected.

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[1]*These traps for electrons are assumed to be neutral when occupied and positive charged when empty (i.e., they are donors).*

2. Model of the current mechanisms in Pt/Ba$_x$Sr$_{1-x}$TiO$_3$/Pt structure.

2.1. Poole-Frenkel mechanism. As it is noted above, if the ferroelectric film contains traps for electrons (conditioned by oxygen vacancies) and is under the simultaneously acting electric field and electron beam irradiation, and, if the adsorbed energy of the trapped electrons from the electron beam is enough to release from traps, the *I-V* characteristics would be exhibited not only on applied voltage dependence, but be changed under the electron beam intensity. For the classic interpretation, in the case of only the applied field in thin film it is expected that Poole-Frenkel emission will "start" dominant up to 1 *MV/cm*. In this case, it is expected that Poole-Frenkel emission can take place for the low electric field too due to the electron beam. Generally, Poole-Frenkel effect [10, 11] describes the lowering of the potential barrier for carrier thermoemission from a localized electronic state as a result of composition of its attractive, Coulomb-like potential that is inversely proportional to the distance, *r*, and of the potential of a uniform external electrical field, *F* (see Fig. 2). For this phenomenon to occur, the defect must acquire a net charge upon the emission of the carrier, i.e. in p-type material Poole-Frenkel effect is expected to occur for acceptor traps and in n-type material – for donor traps. For trap states with Couloumb potential, the charge transport is governed by the Frenkel-Poole emission which is very similar to the Schottky emission [1-12]. It is necessary to note, that while in Schottky mechanism, the lowering of the barrier occurs uniformly for all the directions of the carrier motion in the hemisphere centered in the direction of the field, i.e. the probability of escape is enhanced by the same factor $exp\left(\beta_S E^{\frac{1}{2}}/kT\right)$ for all the attempted directions of escape, the situation for Poole-Frenkel effect is more complicated. In this case, it is necessary to take into account the effect of the angle between the direction of escape and the direction of the field *E*.
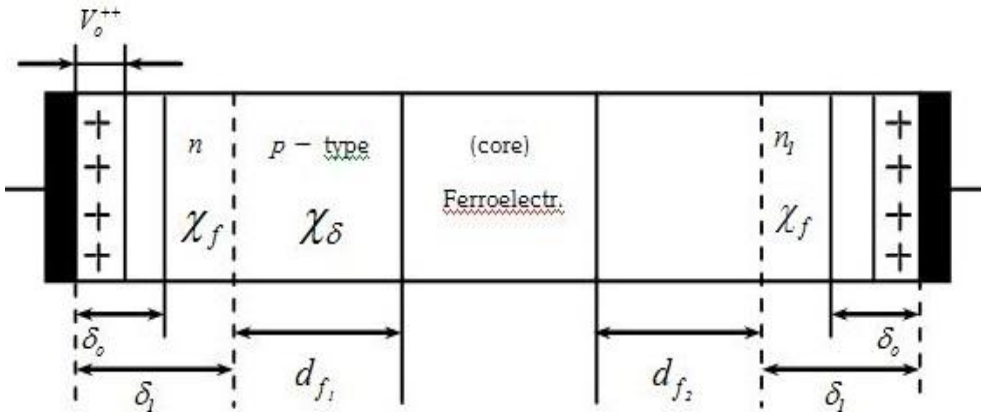

Fig.1. 1D model of a thin film Pt/BSTO/Pt structure [18]

The obtained expression (16) is based on the model, on which the maximum barrier lowering $\left\{\beta_{PF}[E(x)]^{\frac{1}{2}}\right\}$ occurs only in one particular direction in space, all other directions have to overcome a higher barrier.

Fig.2b helps to analyze the relationship between trapped electrons due to Poole – Frenkel effect. Arrows indicate the possible mechanisms of electron emission: thermal ionization over the lowered barrier (PF effect), direct tunneling (DT) into the conduction band (CB), and phonon assisted tunneling (PAT) [10,11].

Assume that a ferroelectric film contains traps for electrons, which have E_{nt} energy levels below the conductance bands. The electron (donor type) trap densities are denoted by N_{td} (Fig.2).
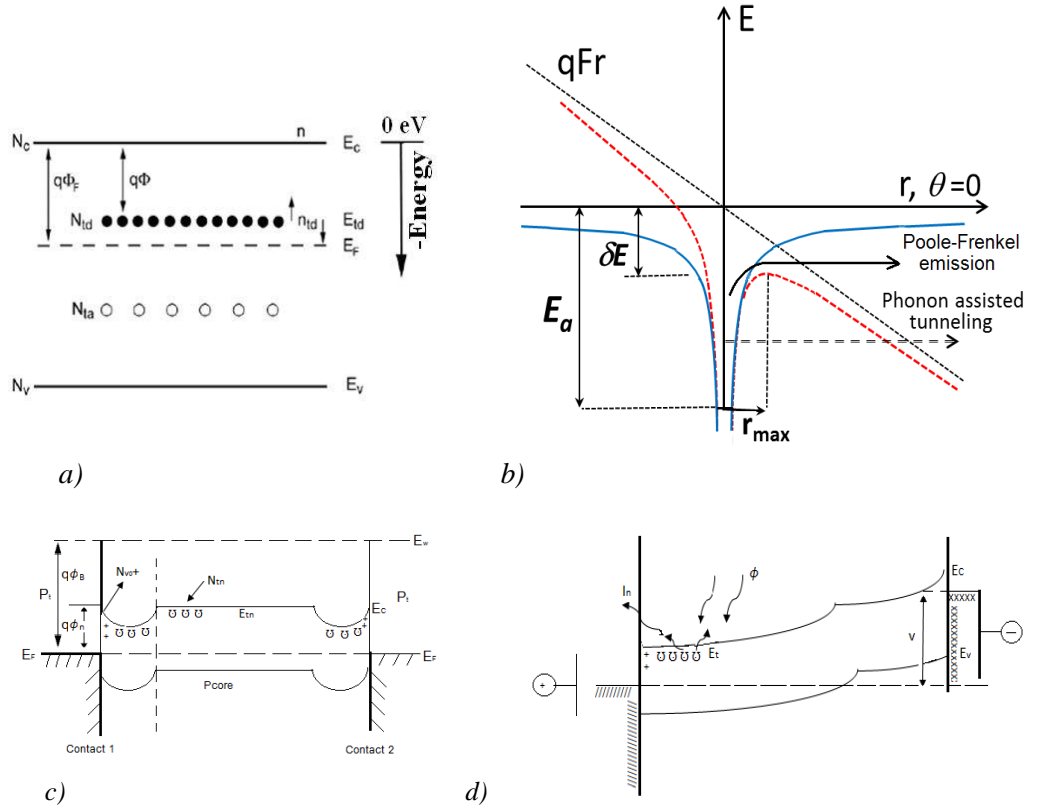

Fig.2. Schematics of trap levels (one-dimensional) for electrons (a), lowering of the barriers by δE under the applied high field, F, [10,11] (b), under equilibrium condition (c) and at irradiation of electron beam(d)

Assume that free electrons in the conduction band of the ferroelectric are a result of only oxygen vacancies and neglecting the intrinsic free electrons in ferroelectrics.

Let's consider that the oxygen vacancies near the metal contact is N_{vo} which act as "donors" and its average energy depth in respect to the edge of the ferroelectric conductance band (E_c) is E_t.

Assume, that the concentration of oxygen vacancies conditioned by the trap density/unit energy is:

$$N_t \cong N_{v_0} exp\left(\frac{E_t - E_c}{kT}\right).$$

Then, for the concentration of ionized "donors", i. e, for the concentration of free electron, we will have:

$$N_{v_0^+} = n_{c_0} \approx N_{v_0}\left[1 - \frac{1}{1+\frac{1}{g}exp\left(\frac{E_t - E_F}{kT}\right)}\right], \tag{1}$$

where g is the ground-state degeneracy for donor levels (g=2), k is the Boltzmann's constant, T is the absolute temperature, E_F is the Fermi level energy.

On the other hand, using the well-known expression for free electron concentration:

$$n_{c_0} = N_c exp\left(\frac{E_F - E_c}{KT}\right) \tag{2}$$

for n_{c_0} we can obtain:

$$n_{c_0} \cong \frac{N_c}{4} exp\left(\frac{E_t - E_c}{kT}\right) * \left\{\sqrt{1 + \frac{8N_{v_0}}{N_c exp\left(\frac{E_t - E_c}{kT}\right)}} - 1\right\}, \tag{3}$$

where N_c is the effective density of states in the conduction band.

For the concentration of trapped electrons at thermal equilibrium we will have:

$$n_{t_0} = N_{v_0} - N_{v_0^+} = \frac{1}{1 + 0.5exp\left(\frac{E_t - E_F}{kT}\right)} = \frac{1}{1 + 0.5exp\left(\frac{E_t}{kT}\right) * \frac{N_c}{n_{c_0}} exp\left(-\frac{E_c}{kT}\right)} =$$

$$= \frac{n_{c_0}}{n_{c_0} + 0.5N_c exp\left(\frac{E_t - E_c}{kT}\right)}, \tag{4}$$

Now, assume that in case of the metal-ferroelectric-metal structure, the electric field and, simultaneously the electron beam excitation (irradiation) is applied. It is assumed that under the influence of the electric field and the electron beam, irradiation will take place. Poole-Frenkel emission of electrons from oxygen vacancies condition the trap levels to the conduction band of ferroelectric (Fig. 2). It means that the escape rate of electrons from trap levels to the conduction band depends not only on the electric field (as in usual Poole-Frenkel effect) but also depends on the electron beam irradiation flux intensity Φ (electr./cm²) if the absorbed energy of trapped electrons is larger than the $(E_t - E_c)$.

Assume that traps, which exhibit a couloumbic attraction in respect to current carriers (electron in this case) is necessary. Thus traps are assumed to be neutral when

occupied and positively charged when empty (i.e., they are "donors"). An expression for the captured electrons $n_t(x)$ under the steady condition (that is under the simultaneously applied electric field and electron beam irradiation) may by obtained by equating the rate of electron capture from the conduction band (r_1) to the rate of electron release from traps (r_2). Considering an infinitesimal trap energy range between E_t and E_t+dE_t, the expression for r_1 can be written as:

$$r_1 = n_c N_t (1 - f_n)\sigma v_{th}dE_t,\tag{5}$$

where σ is the capture cross section, v_{th} is the electron thermal velocity, f_n is the occupancy factor which is a function of trap the energy.

The escape rate can be presented as:

$$r_{zn} = N_t f_n v exp\left(-\frac{E_t-E_c}{kT}\right)exp\left(\beta_{pF}E^{1/2}\right)exp(K_n\Phi)dE_t),\tag{6}$$

$$\beta_{pF} = 2\beta_S = 2\left(\frac{q}{kT}\right)\left(\frac{q}{\pi\lrcorner_{op}}\right)^{1/2},\tag{7}$$

where q is the electronic charge, $\lrcorner_{op}$ is the high frequency dielectric permittivity of ferroelectric, β_S is the Schottky slope, β_{pF} is the slope of the $\log I$-$E^{½}$ plot when the Poole-Frenkel mechanism dominates the electronic conduction, $K_n = \frac{dn_c}{d\Phi}\frac{1}{n_{t0}}$ is the rate of the electron escape from the trap center to the conduction band due to the interaction with the electron beam (due to the absorption of corresponding energy from electron beam).

Let's assume that the entrance of electrons (after electric field is applied) from the cathode (metal contact) into the conduction band of the ferroelectric film by Schottky–thermoionic emission is described by

$$J = J_S exp\left(\beta_S E_{(0)}^{1/2}\right),\tag{8}$$

where J_S is a temperature-dependent constant.

The continuity of the steady-state current inside the film requires that J be constant and independent of the position in the film. Thus, neglecting the diffusion since the electric field is assumed to be high, J is given by

$$J = qn_c(x)\mu E(x),\tag{9}$$

where $n_c(x)$ is the free electron density when the electric field and the electron beam irradiation is applied, μ is the electron mobility, which is assumed to be independent of the field strength, E is the electric field.

The electric field distribution in the ferroelectric field can be determined from Poisson's equation:

$$\frac{\partial[\varepsilon_f(E)E(x)]}{\partial x} = \rho(x),\tag{10}$$

where $\rho(x) = q(N_{v0}^+ - n_t - n_c)$, n_t and n_c are the trapped and free electron concentrations, respectively ε_f is the ferroelectric film dielectric permittivity, which is

assumed to be a nonlinear dependence on the electric field [1,19-21],

$$\varepsilon_f(E) = \frac{\varepsilon_0 \varepsilon(0)}{1+AE^2} = \frac{\varepsilon_r}{(1+AE^2)},$$ (11)

where $\varepsilon_r = \varepsilon_0 \varepsilon(0)$, $\varepsilon(0)$ is the ferroelectric dielectric permittivity at zero field, ε_0 is the free space dielectric constant ($8,85 \cdot 10^{-12}$F/m), and A is constant (for example, for SrTiO$_3$ $A \cong 0,45 * 10^{-11} cm^2/V^2$) [1,19-21].

Then, using (11) for (10) we have:

$$\frac{\varepsilon_r}{\beta}\left(1 - \frac{2AE^2}{\beta}\right)\frac{dE}{dx} = \rho(x), \beta = 1 + AE^2.$$ (12)

Integration of Eq.(12) for boundary condition $x=l$, E=E$_0$, where l is the length of the film, gives:

$$\frac{E_0}{1+A \cdot E_0^2} = \alpha l, \quad \alpha = \frac{q\left(N_{v0}^+ - (n_c + n_t)\right)}{\varepsilon_r}.$$ (13a)

From (13), for the free electron concentration we can get:

$$n_c = N_{V0}^+ - n_t - \frac{\varepsilon_r E_0}{ql(1+AE_0^2)}.$$ (13b)

On the other hand, it is well-known [22-26] that the energy ΔE_{eb} which may absorb atoms from the electron beam during the interaction can be expressed as:

$$\Delta E_{eb} \cong \frac{4meM_a}{(me+M_a)^2}E_e,$$ (14)

where E_e is the flighting electron average energy, me is the electron mass and M_a is the atom's mass.

Returning to r$_2$, it is necessary to note, that r$_2$ depends upon the concentration of centers which are occupied by electrons and on the electron relaxation time T$_{rx}$. In our case, when the electric field and an electron beam excitation are applied on the ferroelectric, for T$_{rx}$ we can use the expression:

$$T_{rx} = \left(\frac{1}{\nu}\right) exp[(E_{tn} - \Delta E_{PF} - \Delta E_{bL})/kT],$$ (15)

where ν is the attempt to escape frequency, ΔE_{PF} represents the lowering of the trap barrier height assuming a Poole-Frenkel mechanism, i.e.

$$\Delta E_{PF} = \beta_{pF}kTE^{1/2},$$

and ΔE_{bL} represents a so called "lowering" in the trap barrier height due to the absorption of the trapped electron's energy from the electron beam.

If the irradiated surface of the ferroelectric is S, the intensity of the electron beam flux is Φ (*electr./cm²*), ΔE_{bL} can be expressed as:

$$\Delta E_{bL} = S\Phi \frac{\Delta n_c}{nt} \Delta E_{eb} = K_n \Phi,$$ (16)

where $K_n = S\frac{\Delta n_c \Delta E_{eb}}{kTnt}$, and Δn_c is the fraction of the trapped electrons which has release to the conduction band.

Equating r$_1$ to r$_2$ and using Eq's (5)-(8), the occupancy factor f_n can be expressed as:

$$f_n = \left\{ 1 + q\, \frac{v\,exp\left(-\frac{\Delta E_t}{KT}\right)exp\left(\beta_{pF}E^{1/2}\right)exp(K_n\Phi)\mu E}{J_s\,exp\left(\beta_s E_{(0)}^{1/2}\right)} \right\}^{-1}. \tag{17}$$

An expression for $n_t(x)$, the total number of the trapped electrons after the electric field and electron beam irradiation, then it follows:

$$n_t(x) = \int_0^{E_{tn}} f_n\, N_{v_0}\, dE_t = \frac{N_{V0}E_t}{(1+\gamma_3)kT}, \tag{18}$$

where $\gamma_3 = \frac{v\,exp\left(\beta_{pF}E^{1/2}\right)exp(K_n\Phi)q\mu E}{\sigma v_{th}J_s\,exp\left(\beta_s E_{(0)}^{1/2}\right)}$ and it is assumed that $\gamma_3 \gg \frac{E_{tn}}{kT} \gg 1$.

Determining the free electron concentration for the condition:

$$n_c = N_{V0}^+ - n_t - \frac{\varepsilon_r E_0}{ql(1+AE_0^2)},$$

finally, for the J we will have:

$$J\left[\frac{A}{cm^2}\right] = q\mu E_0 \left\{ N_{V0}^+ - n_t - \frac{\varepsilon_r E_0}{ql(1+AE_0^2)} \right\}. \tag{19}$$

With the increase of the electric field (E), electron beam intensity, Φ, the concentraction of the trapped electrons, n_t, is decreased. It means, that the concentration of free electrons is increased which, in turn must lead to an increase in the free electron concentration and consequently an increase in J for other equal conditions.

Conclusions. As it follows from expressions (18) and (19), with the increase of electric field (E), electron beam intensity, Φ, the concentraction of the trapped electrons, n_t, is decreased. It means, that the concentration of free electrons is increased which, in its turn, must lead to an increase in the free electron concentration and consequently to the increase of J for other equal conditions.

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ՆՈՒՐԲ ԹԱՂԱՆԹԱՅԻՆ Pt/Ba$_x$Sr$_{1-x}$TiO$_3$/Pt ԿԱՌՈՒՑՎԱԾՔՆԵՐԻ I-V ԲՆՈՒԹԱԳՐԵՐԸ՝ ԷԼԵԿՏՐՈՆԱՅԻՆ ՓՆՋՈՎ ՃԱՌԱԳԱՅԹԵԼԻՍ

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Տեսականորեն հետազոտվել են ֆերոէլեկտրական Pt/Ba$_x$St$_{1-x}$TiO$_3$/Pt (Pt/BSTO/Pt) բարակ թաղանթային կառուցվածքներում կորստյան հոսանքները, երբ թաղանթը ենթարկվում է էլեկտրոնային փնջի ճառագայթման: Ցույց է տրվել, որ եթե էլեկտրոնային փնջից թվածնի վականսիաներով պայմանավորված թակարդային մակարդակներում գտնվող էլեկտրոնների կլանած էներգիան մեծ է այդ մակարդակներից հաղորդական գոտի առաքման ակտիվացիայի էներգիայից, ապա տեղի կունենա էլեկտրոնների՝ Շոտկիի արգելքից ջերմա/էլեկտրական և Պուլ-Ֆրենկելյան միաժամանակյա առաքում:

Առանցքային բառեր. կորստյան հոսանք, ֆերոէլեկտրիկ, գրավման կենտրոն, Շոտկիի արգելք, Պուլ-Ֆրենկելյան առաքում, թվածնի վականսիա:

I-V ХАРАКТЕРИСТИКИ ТОНКОПЛЕНОЧНЫХ Pt/Ba$_x$Sr$_{1-x}$TiO$_3$/Pt СТРУКТУР ПОД ВОЗДЕЙСТВИЕМ ЭЛЕКТРОННОГО ЛУЧА

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Теоретически исследованы токи утечки в сегнетоэлектрических Pt/Ba$_x$St$_{1-x}$TiO$_3$/Pt (Pt/BSTO/Pt) тонкопленочных структурах в случае, когда пленка подвергается воздействию электронного луча. Показано, что если от электронного луча поглощенной энергии электронов больше, чем энергии активации эмиссии из ловушечных уровней в зону проводимости, то одновременно происходят пуль-френкельная и температурная/электрическая (из барьера Шоттки) эмиссии электронов.

Ключевые слова: ток утечки, сегнетоэлектрик, центр захвата, барьер Шоттки, пуль-френкельная эмиссия, кислородная вакансия.