I-V Characteristics of Pt-Ba_{0.25}Sr_{0.75} TiO₃-Pt Thin Films with Oxygen Vacancies

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Received 25 March 2016

Abstract. A new model for calculation of leakage/injection current in ferroelectric perovskite (BST, PZT) metal-ferroelectric-metal structure thin film is considered under the assumption that: a) a high concentration of oxygen vacancies is presence in the interfaced region of metal-ferroelectric contacts, which creates corresponding electron trap levels in the band gap of ferroelectric, and b) the dielectric permittivity of ferroelectric materials have nonlinear dependence on applied electric field. The results of proposed model are compared with the experiment and a good agreement was received.

Keywords: ferroelectric, leakage/injection current, Pool-Frenkelemission, oxygen vacancy

1. Introduction

Interest in ferroelectric thin films has been considerable over the last 20 years, driven by the possibility of using them for non-volatile memory applications and new microelectromechanical systems (MEMS), for ferroelectric FETs, solid-oxide fuel cell (SOFC) electrodes, voltage-tunable capacitors, microwave electronic components, Film Bulk Acoustic Wave Resonators, etc. [1-3].

The leakage/injection current in thin ferroelectric film devices has been a subject for extensive studies [4-7]. The main task of these investigations istounderstand and propose methods for the reduction of the negative impact of the leakage currents on the device performance, and sometimes the leakage currents themselves are used as a sensitive diagnostic tool for monitoring material parameters, such as concentration and distribution of defects and quality of interfaces. The magnitude of the leakage/injection current and the shape of the *I*-V curve depend on the conduction mechanism on dielectric process in the ferroelectric and on the difference in the work function between the electrode and ferroelectric, nature and density of the interface states, traps, etc. Currently it is well established that the oxygen vacancies play a major role. The oxygenconcentration is not a constant throughout of the film. It decreases sharply near the metal electrodes [1, 4, 5], approximately 50% of its value in the center (~ 20nm 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 $1.5 \times 10^{28} m^3$ [1,4,5], while in the 20nm thick 50% depleted (by oxygen) interface layer it is $(3 \times 10^{20} / m^2)$. The missing oxygen results in an image charge on Pt electrode. The surface density of the changes on Pt electrode is 3×10^{16} oxygen/cm² [3]. However, not all of the 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 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 might be explained by conductivity associated with the oxygen vacancies [1-8]. The oxygen vacancies are the most mobile in perovskite ferroelectrics [1-3,9,10]. The experimental results

of both monocrystalline and polycrystalline BSTO at high temperatures [11] shows that the conductivity of undoped polycrystalline titanate oxides depends 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: $O_0 \leftrightarrow \frac{1}{2} O_2(gas) + V_0^{"} + 2e^{-}$. An interstitial oxygen atom can form different types of defects $x = O^0$, O^- , O^{2-} , which means, that triplet states of oxygen correspond three different energies in the energy band of ferroelectric film. Investigation of the electrical and optical properties of deep-level SrTiO₃[1-4,8,12-15] thin film grown by metal-organic chemical vapor deposition show, that dominant defects in the SrTiO₃ consist of a series of deep-level trapping states with energies in the range of $E_v+2.4eV$ to $E_v+3.15eV$ and a series of shallower traps near the conduction band edge in the range of $E_c - E_m = 0.06 \div 0.4 eV$. These dominant electron traps were attributed to oxygen vacancy or iron transition – metal/oxygen vacancy defects. The defect concentration ranges from 10^{14} to 10^{18} cm^3 in the as-grown film. Even this concentration estimated $10^{20} \div 10^{22} cm^3$ [1-3, 8, 12-16]. Since the interface with electrodes is poor with O^{2-} , these interfacial layers are n-type, while internal region is p-type with a local n-type regions. Oxygen vacancies act as donors 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 [12-16]. It is assumed that the donor state is strongly localized around the nearest titanium ions. In the neutral state, the donor level is double occupied, and there is a reduced repulsive interaction between the vacancy and neighboring cations. 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 [17,18]. The interfacial built-in electric fields associated with the trapping centers and oxygen vacancies results in changes the interfacial permittivity of the films [19]. It is worthwhile to notice that the oxygen vacancies are not to be only main defect of ferroelectric films [1-4,7,10,12-16]. The Ba,Sr vacancies in BSTO result in shallow acceptor levels. Dopants also result in levels in the bandgap. Nb gives a shallow level, while Mn, Gr, and Fe give levels near midgap. Pt gives a deep level also near midgap [1-6, 12-15, 20].

Thus, for low density of oxygen vacancies, the undoped ferroelectric titanates films are considered to have slightly p–type conductivity due to the background impurities (i.e. $[Na^+ \text{ for }Pb^{+2}, Fe^{+3} \text{ for }Ti^{+4})$). In fact, the Kelvin probe study [20,21] haveproved that in *PtBaTiO*₃ thin film structure the work function changes from 2.5 $\pm 0.3 \text{ eV}$ for surfaces to 4.4 $\pm 0.4 \text{ eV}$ for the bulk material.

1. Nevertheless, the above-mentioned studies have been performed experimentally. In these experimental studies, the purpose is to detect and verify the injection properties of contacts, the probability of injection mechanisms, (SCL, Schottky, Tunnel, Fowler-Nordheim, Pool-Fraenkel, etc.), the presence of oxygen vacancies (as the inevitable defects in ferroelectric materials) and their impacton various properties of ferroelectric materials.

2. However, there are relatively few studies in which the electrophysical causes of these injection current formation, hysteresis effects in *I-V*, *C-V*, ε -V dependencies, the phenomenon of fatigue, and other phenomena, as well as the connection between those phenomena with nonlinear dependence of dielectric permittivity of electric field, the concentration of deep "traps" level (due to oxygen vacancies and their energy distribution in the bandgap), the phenomenon of charge carriers ionization due to Pool-Frenkel mechanism, etc are theoretically stated in details.

For design and characterization of ferroelectric thin-film based devices, more realistic is the manner where taken into account: a) the presence of oxygen vacancies as the most mobile and

abundant defects in perovskite ferroelectrics; b) the nonlinear dependence of dielectric permittivity of ferroelectric materials on applied electric field. As noted, ferroelectric ceramics and thin films are of widespread technological importance and for that reason, they remain the subject of intense research activity. While their ferroelectric, pyroelectric, piezoelectric, microwave and optoelectronic properties have been well studied experimentally [1-11], according to our knowledge, very little is known so far about the quantitative accountancies of theinfluence of oxygen vacancies and nonlinear dependence of dielectric permittivity of ferroelectric materials on applied electric field to above mentioned *I-V* characteristics of ferroelectric based active and passive components.

It is wellknown, that the theoretical basics of the study of injection currents in solid-state dielectric and semiconductor physics based on self-solution of current density and Poission's equation system. This system, if equation in common case, is nonlinear and is not possible to solve it analytically and usually it is solved by applying the numerical methods.

In this work, we offer new approaches to solve analytically above mentioned nonlinear equation system, which allowsgetting theoptimal results and easy manner formathematical modeling of the I-V characteristics of ferroelectric thin films.

The present study based on following assumptions.

a) The high concentration of oxygen vacancy is "endowed" ferroelectric to n-type semiconductor properties and the low concentration/no-vacancies ferroelectric core exhibit p-type semiconductor properties. In agile microwave devices based on paraelectric phase ferroelectrics, the oxygen vacancies cause high RF and DC leakages currents, microwave losses and distortion (hysteresis) of C-V, $tan\delta V$, and *I-V* dependences[1--11]. In memory applications, the oxygen vacancies cause fatigue [1-4,22].

b) Under applied DC field, the traps release electrons via Poole-Frenkel mechanism and become charged[19]. Due to the change of oxygen vacancies "conditioned" trapped deep electrons occupation (distribution, f(V)) function in films (ceramics) are formed new high electric field polarized includes. The eclectic field of a point charge polarizes the crystal locally, reducing its permittivity which in turn brings to hysteresis behaviour in C(V) and dielectric constant, as well as can impact on I(V) characteristic[19].

c) The dielectric permittivity of ferroelectric materials has nonlinear dependence on applied electric field: $\varepsilon(E, r) = \varepsilon(0)(1 + AE^2)^{-1}$, where $A = 3\beta[\varepsilon_0\varepsilon(0)]^3$ and $\varepsilon(0)$ is the permittivity at zero bias, ε_0 is the vacuum dielectric constant. For example, for SrTiO₃, $\beta = 8 \times 10^9 \text{V} \cdot \text{m}^5/\text{C}^3$, $\varepsilon(0) = 300$ and $A = 0.45 \cdot 10^{-15} \text{ (m/V)}^2$ [1-4,22].

2. Theory and model description

Let us consider Pt - BST - Pt structure. We assume that before applying of the electric field on BST film, the electrical activity of charged oxygen vacancies is compensated by the trapped electrons or corresponding ionized acceptors connected, for instance, with the film non-stochiometry. When external field is applied and increased its magnitude, the part of oxygen vacancies is additionally ionized due to de-trapping of electrons via Pool-Frenkel emission [19,23-26].

The I-V characteristic of the proposed model is considered atneglecting the diffusion currents associated with the charge concentration gradients. In the present analysis, the voltage applied to the structure is assumed to be greater than the voltage necessary to realize the electrons from oxygen vacancies conditioned trap levels to the conduction band. A carrier released from a trap is expected to move in the electric field so, that the re-trapping process may be neglected.

We also assume that ferroelectric film contains traps for electrons, which have E_{nt} energy levels below the conductance bands. The electron trap densities are denoted by N_{tn} . Let's concentration of free electrons (including injected) at the presence of the field is n_c and that traps can exchange electrons onlywith conductance band through the Pool-Frenkel mechanism.

The kinetic equation of an electron change in trap levels:

$$\frac{\partial n_t}{\partial t} = S_n (N_t - n_t) n_c - (P_e + P_F) n_t$$
(1)

where $S_n = \langle V_{th} \rangle \sigma$, $v_{th} = (3kT/m^*)^{1/2}$ is the electron thermal velocity, σ -is the capture cross section of trap, nt-is the concentration of trapped electrons, P_e and P_{pF} are the probabilities of thermal and Pool-Frenkel emission, respectively.

At equilibrium,

$$\left(\frac{\partial n_t}{\partial t}\right) = 0, \quad n_{t0} = \frac{N_t n_0}{n_0 + \gamma(E_t)}, \quad \gamma = \frac{Nc}{g} \exp(E - F_0 / KT),$$

where n_{t0}-is the equilibrium concentration of trapped electrons, N_c is the conductance band effective density of states, g is the degeneracy factor (g=2)[25], E_t-is the energy dept of trap in respect of conductance band edge E_c (E_c=0), F_0 -is the Fermi energy.

Linearizing the current density equation though the film:

$$j = q\mu n_0 E_0 + qD\left(\frac{dn_0}{dx} + \frac{dn_{t0}}{dx}\right) + q\mu n_{t0} E_0 e^{\frac{-E_t - \beta^* E_0^{1/2}}{kT}},$$
(2)

we assume that $\beta^* \equiv q \left(\frac{q\beta}{\pi\varepsilon_{\Gamma}}\right)^{1/2} (\beta^* \text{ is the slope of the logI-} E^{\frac{1}{2}} \text{ plot when the Pool-Frenkel})$

mechanism dominates the electronic conduction or β^* is the trap potential well height lowering), Dis the diffusion coefficient of electrons, q and μ are the electron charge and mobility, respectively. The third component of Eq's (2) is the Pool-Frenkel mechanism conditioned current density. Neglecting the diffusion component of the current, we obtain:

$$j = q\mu n_0 E_0 + q\mu n_0 b E_0 a_1 \left(1 + \frac{E_0^{-1/2} q^{-3/2} \left(1 + A E_0^{-2}\right)^{1/2}}{kT \left(\pi \varepsilon_{\Gamma}\right)^{1/2}} \right) \cong q\mu n_0 E_0 \left(1 + b a_2\right)$$
(3)
$$a_2 = b \exp\left(-\frac{E_t - F_0}{kT}\right) \left(1 + \frac{E_0^{-1/2} q^{-3/2} \left(1 + A E_0^{-2}\right)^{1/2}}{kT \left(\pi \varepsilon_{\Gamma}\right)^{1/2}} \right), \quad a_1 = \exp\left(-\frac{E_t - F_0}{kT}\right), \quad b = \frac{N_t}{\gamma(E_t)}$$

After differentiation of Eq. (3), from the $\frac{dj}{dx} = 0$ condition, for the first approximation, assuming that

$$\beta^* \equiv q \left(\frac{q\beta}{\pi \varepsilon_{\Gamma}}\right)^{1/2} << 1$$
, we obtain:

$$n_0 \frac{dE_0}{dx} = -E_0 \frac{dn_0}{dx}.$$
 (4)

Substituting the expression $\frac{dE_0}{dx}$ into the $n_0 \frac{dE_0}{dx} = -E_0 \frac{dn_0}{dx}$, for the n_0 we can get:

$$n_0 = n_{C0} \exp\left[-\frac{A \cdot j(1+b)x}{\varepsilon_{\Gamma} b_2}\right]$$

Then, substituting the expression of n_0 into the Poission's equation,

$$\varepsilon(E_0)\frac{\partial E_0}{\partial x} + E_0\left(-\frac{2AE_0\varepsilon_{\Gamma}}{\beta^2}\right)\frac{\partial E_0}{\partial x} = q(n_0 + n_{t0}), \quad \frac{dE_0}{dx} = -\frac{qn_0(1+b)AE_0^2}{\varepsilon_{\Gamma}}$$

for the E_0 gives:

$$E_0 = \frac{-\alpha}{\alpha_1} \left(e^{\alpha x} - 1 \right),$$

where $\alpha = \frac{Aj(1+b)}{\varepsilon_{\Gamma}b_2}$, $\alpha_1 = \frac{qn_{C0}(1+b)A}{\varepsilon_{\Gamma}}$.

For the low electric fields, when $AE_0^2 \ll 1$, $\beta \cong AE_0^2 \cong 1$ for *J-V* characteristic of M-F-M structure wereceived:

$$V = \sqrt{\frac{8j(1+b)}{9\varepsilon_{\Gamma}b_2}} \left\{ \left(\frac{\varepsilon_{\Gamma}b_2}{2j(1+b)A} + l \right)^{3/2} - \left(\frac{\varepsilon_{\Gamma}b_2}{2j(1+b)A} \right)^{3/2} \right\},\tag{5}$$

where j-is the current density, V-is the applied voltage, $A = 3\beta[\varepsilon_0\varepsilon(0)]^3 \varepsilon_{\Gamma} = \varepsilon_0\varepsilon(0)\varepsilon(0)$ is the permittivity at zero bias, ε_0 is the vacuum dielectric constant, *l* is the length of ferroelectric film,

$$b_{2} = \mu (1 + b \cdot a_{1}), a_{1} = \exp \left(-\frac{E_{t} - F}{kT}\right), b = \frac{N_{t}}{\gamma(E_{t})}, \gamma(E_{t}) = \frac{N_{c}}{g} \exp(E_{t} - F_{0} / kT).$$

When $\frac{\varepsilon_{\Gamma} \cdot b_2}{2j(1+b)A} \ll l$, Eq. (5) becomes:

$$j = \frac{9V^2 \varepsilon_{\Gamma} b_2}{8(1+b)l^3}.$$

If oxygen vacancies are absent, i.e. $N_t = 0$, and dielectric permittivity does not depend on electric field, i.e., $b_2 = \mu (1 + ba_1) = \mu$, b = 0, we obtain:

$$j = \frac{9V^2 \mathcal{E}_{\Gamma} \mu}{8l^3},\tag{6}$$

which is well known Mott-Henry 2/3 law [25].

For the very high electric fields, if $E_0 \ge 10^6 \text{ V/cm}$, $\left|AE_0^2\right| >> 1$, $\beta \cong AE_0^2$, $1 - AE_0^2 \approx -AE_0^2$, j-V characteristic of M-F-M structure is written as:

$$V = \int_{0}^{l} E_{0} dx = -\frac{\alpha}{\alpha_{1}} \int_{0}^{l} \left(e^{-\alpha x} - 1\right) dx = \frac{(\alpha l)^{2}}{2\alpha_{1}} \cong l^{2} \frac{A^{2} \cdot j^{2}(1+b)^{2} \cdot \varepsilon_{\Gamma}}{2\varepsilon_{\Gamma}^{2} \cdot b_{2}^{2} \cdot q \cdot n_{c0} \cdot (1+b)A}, \quad j = \frac{b_{2}}{l} \left(\frac{2V\varepsilon_{\Gamma} \cdot q \cdot n_{c0}}{A(1+b)}\right)^{1/2}$$
(7)

As it follows from Eq.(7), the jhas $\frac{1}{2}$ law dependence on V which differ from (6) via quantitative coefficient which may be as a consequence of our approximations during solution.

Theoretical calculations havebeen carried out according to (7) at following parameters of ferrofilm and Pt-BST-Pt structure:Ba_{0.25}Sr_{0.75}TiO₃, l = 0.5 cm, ε (0)=300, area $S_o \sim 3 \cdot 10^{-6} cm^2$, A=0.45 $\cdot 10^{-15} (m/V)^2$, $\mu_{no} \cong (10^{-1}....6)cm^2 / Vs$, $\varepsilon_f \approx (10 \div 20)$, $E_{in} = 0.026 \div 0.4 eV$, $n_{c_0} \cong (10^{12} \div 10^{14})cm^{-3}$, $N_t = 10^{16}...10^{19} cm^{-3}$ [1-12,15,19] and T=300K.Theoretical calculations have been compared with the experimental results (Figs.2-5).

3. Experimental results and discussions

The BST films (~100 nm) of Ba_{0.25}Sr_{0.75}TiO₃ composition were prepared by PLD technique, carried out in Aachen University of Applied Sciences, Institute of Nano- and biotechnologies, Campus Juleich, Germany, using targets fabricated via the self-propagating high-temperature synthesis (SHS) method. The process steps of the BST synthesis are described in detail in [27]. The deposition was performed in an oxygen atmosphere (gas flow 30 mL/min, pressure $2 \cdot 10^{-3}$ mbar) using a KrF-excimer laser (Lambda LPX305) with a pulse width of 20 ns and a pulse energy of approximately 1 J per pulse. When using a deposition time of 300 s, an energy density of 2.5 J cm⁻² and a repetition rate of 10 Hz, the BST layer thickness amounted approximately to ~100 nm. Finally, the chip coated with BST was mounted on a printed circuit board (PCB), followed by ultrasonic wire bonding and encapsulation processes. Experiments havebeen carried out on KEITHLEY 6430 Sub-FemtoampSourceMeter.

Fig.1 shows the schematic cross section (a) and the top view (b) of the fabricated Pt-BST-Pt structure. Each of the Pt electrodes had a 3 mm diameter, and thelength, width and thickness of BST were10 mm, 5 mmand ~100 nm, respectively.



Fig.1. The layered structure of Pt - Ba_{0,25}Sr_{0,75}TiO₃-Pt sample(a) and top view photograph (b)

In Fig.2 is presented the calculated (according to Eq.7) and the experimental *I-V* dependencies of $Pt-Ba_{0,25}Sr_{0,75}TiO_3$ -Ptstructure. Fig.3 depicts the theoretical *I-V* dependence of examined structure at different concentration of oxygen vacancies, N_t . In Fig.4 and Fig.5 are presented the theoretical dependence of current on energy dept of oxygen vacancies conditioned traps, E_t , at different values

of oxygen vacancies N_t , and dependence of current on oxygen vacancies concentrationat different values of energy dept of oxygen vacancies conditioned traps, E_t , respectively.



Fig.2. Theoretical (blue) and experimental (red) dependencies of J(V) of Pt – Ba₀₂₅Sr_{0,75}TiO₃ – Pt structure (by Eq.7), $(T = 25 \ \mathcal{C}, \mu_0 = 6.0 \text{ cm}^2/\text{Vs}, \text{ E}_t = 0.036 \text{ eV}, n_0 = 6.6 \cdot 10^{12} \text{ cm}^{-3}, N_t = 10^{18} \text{ cm}^{-3})$



Fig.3. Theoretical dependence of J(V) for Pt – Ba_{0,25}Sr_{0,75}TiO₃ – Pt structure at different values of oxygen vacancies concentration N_t (μ_0 =6.0cm²/Vs, E_t=0.036 eV, n_0 =6.6 \cdot 10¹²cm⁻³).



Fig.4. Theoretical dependence of current, J (A/cm²), of Pt – Ba₀₂₅Sr_{0,75}TiO₃– Ptstructure on energy dept of oxygen vacancies conditioned traps , E_t, at different values of oxygen vacancies N_t .

 $(\mu_0=6.0 \text{ cm}^2/\text{Vs}, \text{ V}=10 \text{ V}, n_0=6.6 \cdot 10^{12} \text{ cm}^{-3})$



Fig.5. Theoretical dependence of current, J(A/cm²), of Pt – Ba₀₂₅Sr_{0,75}TiO₃ – Pt structure on oxygen vacancies concentration N_t at different values of energy dept of oxygen vacancies conditioned traps , Et. (μ_0 =6.0cm²/Vs,V=10 V, n_0 =6.6 · 10¹²cM⁻³)

From Fig.2 follows, that there are good agreements between the results of proposed model and experiments. With the increase of oxygen vacancies concentration, N_t (i.e. with the increase of the trap levels captured electrons concentration) the current is increases due to Pool-Frenkel ionization(Fig.3). Thefact that the J(E_t) dependence passing through the minimum (Fig.4, Fig.5), itcan be explained by the following model: at the constant concentration of oxygen vacancies, N_t, i.e. trap centers with the increase of its energy dept, E_t, current is decreases due to dominate of capture rate of the injected charge carriers in respect of escape from those traps. At higher energy dept, when all traps are fully occupied, the escape becomes dominant in respect of captureat the all other equal conditions.

Acknowledgment

The authors are grateful to Prof. M. Schoening, Prof. A. Poghossian and Dr. C. Huckfrom Aachen University of Applied Sciences, Institute of Nano- and biotechnologies, Campus Juleich, Germany, for valuable discussions and support during the preparation of samples. This work was supported by State Committee of Science MES RA, in frame of the research project № SCS 14AR-2f12.

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