

Electrophysical Properties Investigation of $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ Ferroelectric Films in Paraelectric State

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Abstract—Phenomenological analytical expressions relating the ferroelectric film capacity to the coefficients of free energy decomposition in accordance to the degrees of substance polarization in paraelectric state are obtained. High-frequency capacitance-voltage characteristics of metal– $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ –metal structures have been measured at the temperatures above the transition point from ferroelectric phase to paraelectric one. The comparison of theoretical and experimental data has been resulted in plotting the dependences of $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ film polarization on external voltage and determining the values of the parameters of the theory of phase transitions of second kind of Ginsburg–Landau for the objects under investigation.

Keywords: metal–dielectric–metal structures, ferroelectric $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ films, high-frequency capacitance-voltage characteristics, capacity

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1. INTRODUCTION

Based on metal–oxide–semiconductor silicon structures nanoelectronics actually reached the limits of SiO_2 insulating film thickness reduction. It follows from the fact that minimum crystallographic size of two transition layers at the interphase boundaries between Si-substrate– SiO_2 and field electrode–“poly-Si– SiO_2 ” is equal to 16 Å [1]. That's why the researches are undertaken for many years in order to find the alternative to silicon oxide for electronic device usage. One of the priority directions is to use ferroelectric films as insulating layers [2]. Unique physical properties of ferroelectrics (in particular, high and changeable in the external electric field dielectric constant) allow to create based on the planar technology the actually new class of the devices for information storage and processing [3]. Solid solutions of barium-strontium titanate $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ or BSTO appear to be the promising type of ferroceramics for microwave technology. They have ferroelectric properties at room temperature and dielectric constant of their thin films remains sufficiently high in wide temperature range. The investigation of BSTO films has started since the end of the last century [4, 5]. However, there is still no methodology of plotting the data on the coefficients of free energy decomposition by degrees of the order parameter—the dielectric polarization of the substance P —from the measured capacitance-voltage characteristics (CVC). Knowledge of these coefficients is necessary, since it is they that determine the

electrophysical properties of ferroelectric structures. Difficulty mentioned above (the absence of methodology) is caused by spontaneous polarization of the substance in ferroelectric phase due to spatially distributed domains [6]. That's why the expression for the dependence of ferroelectric film capacity on the applied voltage should be based on a three-dimensional averaging of domain polarization over space. Spontaneous polarization is absent in paraelectric phase and as was emphasized in [7] the problem of taking into account the spatial distribution of polarization becomes one-dimensional. In this paper the phenomenological analytical expressions relating the ferroelectric film capacity to the coefficients of free energy decomposition in accordance to the degrees of substance polarization in paraelectric state are obtained; the results of high-frequency CVC of metal–BSTO–metal structures measured are shown and the values of the parameters of the theory of phase transitions of second kind of Ginsburg–Landau are determined for the experimental objects. It is worth mentioned that the coefficients of free energy decomposition are temperature dependent and their values found in paraelectric phase do not match with the values in ferroelectric phase; however, in principle, it is possible to make an extrapolation in ferroelectric range, for example, according to the Curie–Weiss law and get an idea of the parameters below the phase transition temperature.

2. THEORETICAL PART

When presenting the theoretical part we will follow [7] including the notations. In accordance to the theory of phase transitions of second kind of Ginsburg–Landau the free energy when decomposed by the square of polarization to the second term has the following form

$$F = F_0 + \frac{2\pi}{k} P^2 + \frac{A P^4}{4 P_0^2} - EP. \quad (1)$$

Here F_0 is free energy in the absence of polarization (i.e., without electric forces); $\frac{4\pi}{k}$ is the reverse susceptibility; E , P , and D are the averaged on the scales of the crystal cell electric field, polarization and induction, respectively; P_0 is characteristic intrinsic polarization of the crystal cell, $P_0 = \frac{q}{a^2}$, where q is elementary charge, a is the characteristic size of the crystal cell; A is dimensionless coefficient of the order of one. We were interested to determine the parameters k and $\frac{P_0^2}{A}$ from the experimental CVC. Minimum of free energy in equilibrium is achieved under the condition $\left. \frac{\partial F}{\partial P} \right|_{E=\text{const}} = 0$. Therefore,

$$E = \frac{4\pi P}{k} + A \frac{P^3}{P_0^2}, \quad (2)$$

$$D = E + 4\pi P = 4\pi P \left(a + \frac{1}{k} \right) + A \frac{P^3}{P_0^2}.$$

By definition [6], the dielectric constant of the ferroelectric is the following

$$\kappa_{\text{se}} \equiv \frac{dD}{dE} = 1 + \frac{k}{1 + \frac{3Ak P^2}{4\pi P_0^2}}$$

and, therefore, depends on both z coordinate and the voltage between the electrodes V_g [since $P = P(z, V_g)$]. For the phase transition to exist at the Curie temperature T_C , it is necessary for the parameter k to be dependent on the temperature according to the Curie–Weiss law: $k^{-1} = \alpha(T - T_C)$, where α is the constant slightly dependent on the temperature, T is the temperature in degrees Celsius. For spontaneous polarization P_s in ferroelectric phase under $E = 0$ it turns out [6]:

$$P_s = P_0 \left[\frac{4\pi\alpha(T_C - T)}{A} \right]^{1/2}. \quad (3)$$

Let's consider a potential relief in a sample with metallic contacts on the surfaces of a ferroelectric film. A significant difference in the states of ferroelectric being in the form of a thin film or massive sample is the complete depletion of volume by free charge carriers. Due to the large values of dielectric constant for typical concentration of free charge carriers $N =$

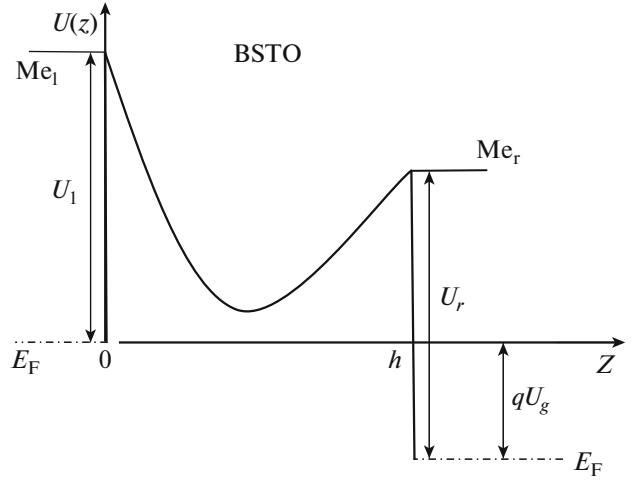


Fig. 1. Zone diagram of a ferroelectric film with metal electrodes; E_F is Fermi level; Me_l and Me_r are the metals left and right of BSTO film; $U(z)$ is the potential of electrons counted from the bottom of the BSTO conduction band; h is BSTO film thickness.

10^{15} cm^{-3} the thickness of Schottky depletion layer near the contact of massive ferroelectric with metal exceeds several microns [8]. As a result, there is a qualitative difference between the film and massive sample: the distribution of polarization in thin sample is inhomogeneous over the thickness at least under relatively low external voltages. Figure 1 shows zone diagram of a thin film metal–BSTO–metal structure. Potential of electrons in ferroelectric near Schottky contacts with metal describes two barrier reliefs which merge into a single line inside BSTO. The dependences of the field and polarization on the coordinate are described by Poisson equation

$$\frac{dD}{dz} = \frac{d}{dz} \left[4\pi P \left(1 + \frac{1}{k} \right) + A \frac{P^3}{P_0^2} \right] = 4\pi q N, \quad (4)$$

and potential is determined from the expression

$$\frac{dU}{dz} = qE = q \left(\frac{4\pi P}{k} + A \frac{P^3}{P_0^2} \right). \quad (5)$$

Boundary conditions to the system of equations (4), (5) are the following: $U|_{z=0} = U_l$, $U|_{z=h} = U_r - qV_g$. Here U_l and U_r are the work functions to BSTO from the left and right metals, respectively; h is ferroelectric film thickness. Let's indicate $P|_{z=0} = P_l$, $P|_{z=h} = P_r$. The solution of the task (4), (5) is written as

$$\begin{cases} P \left(1 + \frac{1}{k} \right) + \frac{A P^3}{4\pi P_0^2} = P_l \left(1 + \frac{1}{k} \right) + \frac{A P_l^3}{4\pi P_0^2} + qNz, \\ U = U_l + \frac{2\pi(k+1)}{k^2 N_d} \left[(P^2 - P_l^2) + \frac{(k+4)(Ak)}{2(k+1)(4\pi)} \right. \\ \left. \times \frac{(P^4 - P_l^4)}{P_0^2} + \frac{1}{(k+1)(4\pi)^2} \frac{(P^6 - P_l^6)}{P_0^4} \right]. \end{cases} \quad (6)$$

Moreover

$$\begin{cases} P_r \left(1 + \frac{1}{k}\right) + \frac{A}{4\pi} \frac{P_r^3}{P_0^2} = P_l \left(1 + \frac{1}{k}\right) + \frac{A}{4\pi} \frac{P_l^3}{P_0^2} + qNh, \\ U_r - qV_g - U_l = \frac{2\pi(k+1)}{k^2 N_d} \left[(P_r^2 - P_l^2) + \frac{(k+4)}{2(k+1)} \right. \\ \left. \times \left(\frac{Ak}{4\pi} \frac{(P_r^4 - P_l^4)}{P_0^2} + \frac{1}{(k+1)} \left(\frac{Ak}{4\pi} \right)^2 \frac{(P_r^6 - P_l^6)}{P_0^4} \right) \right]. \end{cases} \quad (7)$$

Now let's determine the capacity of metal–BSTO–metal structure C . Its expression is the following: $C = S \frac{d\sigma_r}{dV_g}$, where S is the area of the structure,

$\sigma_r = \frac{D_r}{4\pi} = P_r \left(1 + \frac{1}{k}\right) + \frac{A}{4\pi} \frac{P_r^3}{P_0^2}$ is the charge per unit area of the right electrode. From the expressions (7) for the capacity we obtain

$$C = \frac{SkqN}{4\pi\Gamma} \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{P_l^2}{P_0^2} \right] \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{P_r^2}{P_0^2} \right], \quad (8)$$

$$\begin{aligned} \Gamma = & P_r \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{P_l^2}{P_0^2} \right] \left[1 + \frac{(k+4)}{(k+1)} \left(\frac{Ak}{4\pi} \right) \frac{P_r^2}{P_0^2} \right. \\ & \left. + \frac{3}{(k+1)} \left(\frac{Ak}{4\pi} \right)^2 \frac{P_r^4}{P_0^4} \right] - P_l \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{P_r^2}{P_0^2} \right] \\ & \times \left[1 + \frac{(k+4)}{(k+1)} \left(\frac{Ak}{4\pi} \right) \frac{P_l^2}{P_0^2} + \frac{3}{(k+1)} \left(\frac{Ak}{4\pi} \right)^2 \frac{P_l^4}{P_0^4} \right]. \end{aligned} \quad (9)$$

The dependence of capacity on the voltage is determined by the functions $P_l(V_g)$, $P_r(V_g)$, which, in turn are determined from equalities (7). Maximum value of capacity is achieved at $U_r - qV_g - U_l = 0$ and deviation of $C(V_g)$ curve from symmetry with respect to the change in voltage sign is due to the difference in work functions from the metals. In the point of maximum

$P_r = -P_l = P_m$, $P_m \left(1 + \frac{1}{k}\right) + \frac{A}{4\pi} \frac{P_m^3}{P_0^2} = \frac{qNh}{2}$ and the expression for capacity becomes the following

$$C_{\max} = \frac{S(k+1)}{4\pi h} \times \frac{\left[1 + \frac{Ak}{4\pi(k+1)} \frac{P_m^2}{P_0^2} \right] \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{P_m^2}{P_0^2} \right]}{\left[1 + \frac{(k+4)}{(k+1)} \left(\frac{Ak}{4\pi} \right) \frac{P_m^2}{P_0^2} + \frac{3}{(k+1)} \left(\frac{Ak}{4\pi} \right)^2 \frac{P_m^4}{P_0^4} \right]}. \quad (10)$$

When the value of P_m is small as compared with characteristic intrinsic polarization of crystal cell, i.e.,

$\frac{AP_m^2}{4\pi P_0^2} \ll 1$, then $P_m \cong \frac{qNhk}{2(k+1)}$; and when this inequality

is enhanced up to $\frac{AkP_m^2}{4\pi P_0^2} \ll 1$, then the expression

(10) goes into the formula of flat capacitor: $C_{\max} \cong \frac{S(k+1)}{4\pi h}$. The value of the parameter k is high especially

near Curie temperature, therefore, even under relatively small voltages, with the increase of the external field the contribution of non-linear terms like

$\left(\frac{Ak}{4\pi} \right) \frac{P_r^2}{P_0^2}$, $\left(\frac{Ak}{4\pi} \right) \frac{P_l^2}{P_0^2}$ to multiplier Γ begins to affect (see the expression (9)). It results in drop of the capacity value with voltage, and the dependence $C(V_g)$ near the extreme point $qV_g = U_r - U_l$ may be written in the form

$$C = \frac{C_{\max}}{1 + \gamma(U_r - qV_g - U_l)^2}, \quad (11)$$

where $\gamma = \frac{1}{2q^2\Gamma} \frac{d^2\Gamma}{dV_g^2} \Big|_{U_r - qV_g - U_l = 0}$. In general, the expression for parameter γ has a bulky look but it is simplified

under the conditions $\frac{AP_m^2}{4\pi P_0^2} \ll 1$ up to the form

$$\begin{aligned} \gamma \cong & \frac{3Ak^3(k+3)}{64\pi^3(k+1)q^2h^2} \frac{1}{P_0^2} \\ & \times \left[1 + \frac{(k+4)}{(k+1)} \left(\frac{Ak}{4\pi} \right) \frac{P_m^2}{P_0^2} + \frac{3}{(k+1)} \left(\frac{Ak}{4\pi} \right)^2 \frac{P_m^4}{P_0^4} \right]. \end{aligned} \quad (12)$$

Polarization in the film increases with the external voltage increase and there comes a moment when the inhomogeneity of the field in a ferroelectric can be neglected: $|P_r - P_l| \ll |P_r| + |P_l|$. Let us introduce the

value of average polarization: $\bar{P} = \frac{1}{2}(P_r + P_l)$. As follows from (7)

$$\begin{cases} (P_r - P_l) \cong \frac{qNhk}{(k+1)} \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{\bar{P}^2}{P_0^2} \right]^{-1} \\ \bar{P} \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{\bar{P}^2}{P_0^2} \right]^{-1} \left[1 + \frac{(k+4)}{(k+1)} \left(\frac{Ak}{4\pi} \right) \frac{\bar{P}^2}{P_0^2} \right. \\ \left. + \frac{3}{(k+1)} \left(\frac{Ak}{4\pi} \right)^2 \frac{\bar{P}^4}{P_0^4} \right] \cong \frac{k}{4\pi qh} (U_r - qV_g - U_l). \end{cases} \quad (13)$$

Condition for implementation of the homogeneous polarization mode of the ferroelectric film is written in the form of inequality

$$|U_r - qV_g - U_l| \gg \frac{4\pi q^2 N h^2}{(k+1)} \left[1 + \frac{3Ak}{4\pi(k+1)} \frac{\bar{P}^2}{P_0^2} \right]^2$$

$$\times \left[1 + \frac{(k+4)}{(k+1)} \left(\frac{Ak}{4\pi} \right) \frac{\bar{P}^2}{P_0^2} + \frac{3}{(k+1)} \left(\frac{Ak}{4\pi} \right)^2 \frac{\bar{P}^4}{P_0^4} \right].$$

$$\Gamma \cong (P_r - P_l) \left[1 + \frac{(k+3)}{(k+1)} \left(\frac{3Ak}{4\pi} \right) \frac{\bar{P}^2}{P_0^2} + \frac{(2k+3)}{(k+1)^2} \left(\frac{3Ak}{4\pi} \right)^2 \frac{\bar{P}^4}{P_0^4} + \frac{1}{(k+1)^2} \left(\frac{3Ak}{4\pi} \right)^3 \frac{\bar{P}^6}{P_0^6} \right],$$

As it appears from (9) the value Γ is proportional to $|P_r - P_l|$ in the mode under study and the correlation itself may be rewritten in the form

and for the capacity from (8) taking into account the first equality of (13), it turns out that

$$C \cong \frac{S(k+1)}{4\pi h} \frac{\left[1 + \frac{3Ak}{4\pi(k+1)} \frac{\bar{P}^2}{P_0^2} \right]^3}{\left[1 + \frac{(k+3)}{(k+1)} \left(\frac{3Ak}{4\pi} \right) \frac{\bar{P}^2}{P_0^2} + \frac{(2k+3)}{(k+1)^2} \left(\frac{3Ak}{4\pi} \right)^2 \frac{\bar{P}^4}{P_0^4} + \frac{1}{(k+1)^2} \left(\frac{3Ak}{4\pi} \right)^3 \frac{\bar{P}^6}{P_0^6} \right]}. \quad (14)$$

When inequalities $\frac{AkP_m^2}{4\pi P_0^2} \ll 1$, $\frac{A\bar{P}^2}{\pi P_0^2} \ll 1$ are satisfied the expressions for the average polarization (13) and for capacity (14) are simplified and take the form

$$\bar{P} \left[1 + \frac{(k+4)}{(k+1)} \left(\frac{Ak}{4\pi} \right) \frac{\bar{P}^2}{P_0^2} \right] \quad (13a)$$

$$\cong \frac{k}{4\pi q h} (U_r - V_g - U_l),$$

$$C \cong \frac{C_{\max}}{\left[1 + \frac{(k+3)}{(k+1)} \left(\frac{3Ak}{4\pi} \right) \frac{\bar{P}^2}{P_0^2} \right]}. \quad (14a)$$

In conclusion of the theoretical part, we emphasize once again that the functional dependence of the capacity of a thin ferroelectric film on the applied voltage is associated with an ever-increasing contribution to the free energy of its components with fourth and higher degree of polarization.

3. EXPERIMENTAL TECHNIQUE

Metal–BSTO–metal structures were prepared on silicon substrate with platinum sublayer. Ferroelectric $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ film of the thickness $h = 350$ nm was formed by high-frequency sputtering of the polycrystalline target in oxygen atmosphere on Plasma-50SE (Russia) equipment. The substrate temperature was 620°C during the process. The upper nickel electrode was formed on the ferroelectric film surface by electron-beam deposition through the shadow mask on A700QE/DI12000 (Germany) equipment. Electrode size was 2.7×10^{-4} cm², thickness was 0.1 μm . The details of experimental technique, sputtering and deposition equipment are described in [9, 10]. Measurements of CVC of Ni–BSTO–Pt structures were

accomplished on the measuring automated stand [11] equipped by the precision meter LCR Agilent E4980A, portable computer with modified software and special chamber with the heating table with thermo stabilization. The bias voltage V_g was applied to the sample within the range from -15 V up to $+15$ V in steps of 0.25 V, the amplitude and the measuring signal frequency were 25 mV and 100 kHz, respectively. The dynamic scan speed was 0.75 V/s and the data from 3 dots per second were recorded to computer memory.

4. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2 shows the typical CVC of Ni–BSTO–Pt structure measured at 121°C . Curie temperature of the massive $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ sample lies in the range of 60 – 80°C [12, 13] (the values given in these reviews for our compound differ in 20 units). CVC maximum is achieved at the voltage $V_{g\max} = -0.83$ V and differs insignificantly from the difference in work functions from nickel and platinum -0.5 eV [14]. The possible reason of deviation is the delay effects: CVC curve being measured inversely to the shown in Fig. 2 dynamic scanning direction achieves maximum at $V_{g\max} = -0.28$ V. Numerical estimation of the value $\frac{qNh}{2}$ at free electron concentration $N = 10^{15}$ cm⁻³ leads to 2.5×10^3 V/cm, and for self-polarization of the crystal cell at $a = 5 \times 10^{-8}$ cm the value is $P_0 = 5.8 \times 10^7$ V/cm. Hence, we have $\frac{AkP_m^2}{4\pi P_0^2} \ll 1$ and $C_{\max} \cong \frac{S(k+1)}{4\pi h}$ under the experimental conditions. The value C_{\max} in CVC maximum is equal to 6.3×10^{-10} F, therefore, the value $k \approx 922$ is found at 121°C . In the mode of ferroelectric film homogeneous polarization it is possi-

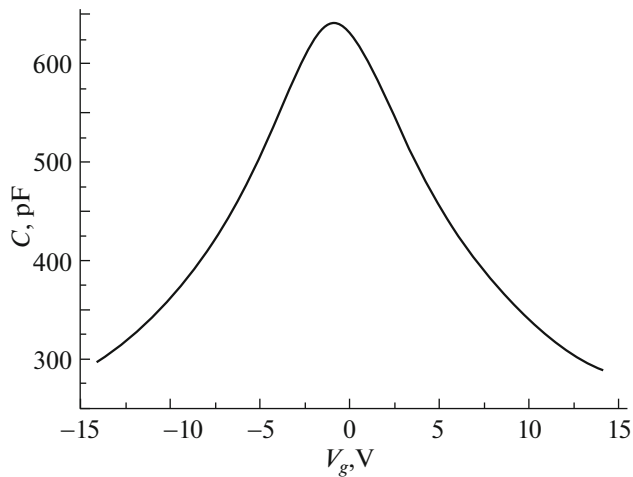


Fig. 2. High-frequency capacity-voltage characteristic of Ni-BSTO-Pt structure measured at 121°C.

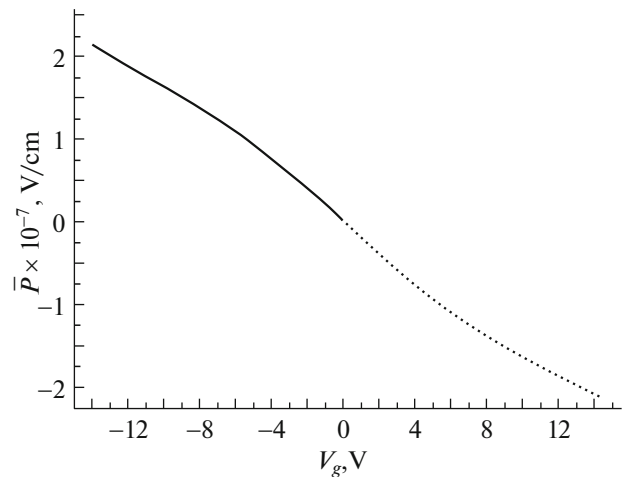


Fig. 3. Dependence of the average polarization of Ba_{0.8}Sr_{0.2}TiO₃ film on the voltage.

ble using the expression (14a) to determine from the experimental curve $C(V_g)$ the relation between quadratic term and voltage: $\frac{(k+3)}{(k+1)} \left(\frac{3Ak}{4\pi} \right) \frac{\bar{P}^2}{P_0^2} = \frac{C_{\max}}{C(V_g)} - 1$. Then, the average polarization is calculated from the equation (13a) as the function of the applied voltage

$$\bar{P}(V_g) = \frac{k}{4\pi h} \left\{ 1 + \frac{(k+4)}{3(k+3)} \left[\frac{C_{\max}}{C(V_g)} \right] \right\}^{-1} (V_{g \max} - V_g).$$

Figure 3 shows the curve $\bar{P}(V_g)$ resulted from these calculations. It can be seen that the nonlinearity of the dependence begins to appear with the voltages in the region of 10 V when the experimental capacity is reduced by almost 2 times. Large values of average polarization (up to 2×10^7 V/cm) are due to the significant value of $k \approx 922$. The experimental curve $\bar{P}(V_g)$ allows to determine the value $\frac{A}{P_0^2}$.

As it follows from (14a)

$$\frac{A}{P_0^2} \cong \frac{h^2}{3(V_{g \max} - V_g)^2} \left(\frac{4\pi}{3} \right)^3 \frac{(k+1)}{(k+3)} \times \left(\frac{C_{\max}}{C} - 1 \right) \left\{ 1 + \frac{(k+4)}{3(k+3)} \left[\frac{C_{\max}}{C(V_g)} - 1 \right] \right\}^2.$$

The dependence of the right part of this expression on the voltage is shown in Fig. 4. It is worth keeping in mind analyzing this curve that it was plotted in the concept of homogeneous distribution of polarization across the sample; vertical lines in Fig. 4 limit within themselves the area where the conditions of homoge-

neity are not satisfied. Under the large voltages the found value $\frac{A}{P_0^2}$ changes with voltage up to 60%. This indicates that the measurement accuracy is not high enough. The experimental values of P_0 were found to be 5–10 times higher than those obtained in numerical estimation (the value of parameter A is of order of one [6]). This occurred due to the fact that in estimates a rather large value was accepted as the characteristic size of the crystalline cell a : 5 Å. Its halving practically equalizes the experimental and evaluative results.

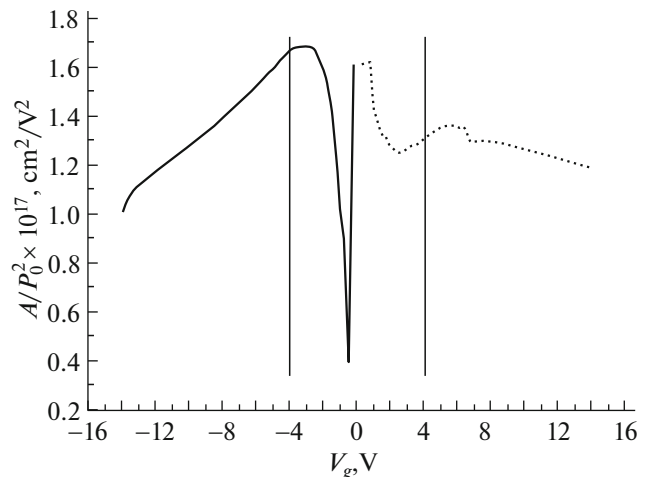


Fig. 4. The parameter $\frac{A}{P_0^2}$ under the different voltages. Two vertical lines limit within themselves the area near the point of maximum capacity of the structure where the polarization cannot be considered as homogeneous.

5. CONCLUSIONS

It is shown that it is possible to identify the dependences of ferroelectric film polarization on the external voltage and to determine the values of the parameters of the theory of phase transitions of second kind of Ginsburg–Landau from the experimental measurements of CVC of metal–BSTO–metal structures having been in paraelectric phase. It is ought to be noted

that in typical experimental conditions $\frac{AkP_m^2}{4\pi P_0^2} \ll 1$ the parameters N and P_m characterizing inhomogeneity of polarization distribution over the film thickness are included in expressions for measured values only in the form of small additives. Therefore, their determination is the task of the other studies, for example, the study of the conductive properties of BSTO films.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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