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## Research Article

# Asymmetry of Polarization Reversal and Current-Voltage Characteristics of Pt/PZT-Film/Pt:Ti/SiO<sub>2</sub>/Si-Substrate Structures

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The characterization of the asymmetries of bipolar charge-voltage and current-voltage loops of polarization reversal and unipolar current-voltage curves for Pt/PZT-film/Pt:Ti/SiO<sub>2</sub>/Si-substrate systems was performed in the dynamic mode. The asymmetry of local deformation-voltage loops was observed by piezoresponse force microscopy. The comparison of the dependences of introduced asymmetry factors for the bipolar charge-voltage and current-voltage loops and unipolar current-voltage curves on drive voltage indicates the interconnection of ferroelectric and electrical space charge transfer asymmetries.

## 1. Introduction

At present, lead zirconate titanate (PZT) film on silicon structures are considered among the best ones for creating elements of Si-integrated converters of various types. However, the practically important electrical characteristics of “metal-PZT film-metal-Si-substrate” systems manifest the well-known set of natural and technological asymmetries.

Earlier [1–3], we reported the results of investigations of polar and poling asymmetries of the set of pyroelectric and related characteristics of Pt/PZT-film/Pt:Ti/SiO<sub>2</sub>/Si-substrate sandwich type structures.

In this paper, we concentrate our attention on the characterization of the complex of the asymmetries of dynamic bipolar charge-voltage (Q-V-) and current-voltage (I-V-) loops and unipolar current-voltage (J-V-) curves as well as bipolar deformation-voltage (X-V-) loop asymmetry. We present the results of investigation of interrelated polarization reversal and current-voltage dynamic asymmetries of Pt/PZT-film/Pt:Ti/SiO<sub>2</sub>/Si-substrate systems.

## 2. Experiment

**2.1. Samples.** The samples under investigation were PZT films prepared by the radiofrequency magnetron sputtering

method. The bottom Pt electrode of 150 nm of thickness with Ti adhesive layer of 10 nm of thickness was deposited on the 350 nm SiO<sub>2</sub> layer on a 350 μm (100) *n*-type Si wafer. Pb(Zr<sub>x</sub>Ti<sub>1-x</sub>)O<sub>3</sub> film with  $x = 0.54$  of 1–2 μm of thickness was deposited on the Pt/TiO<sub>x</sub>/SiO<sub>2</sub>/Si-substrate structure just after its stabilization by annealing treatment at 650–700°C.

The perovskite ferroelectric phase of PZT was obtained by annealing at 625°C for 30 min. Then, a top Pt (150 nm) electrode of ≈1 mm<sup>2</sup> of area was deposited by sputtering.

The sputtering conditions and the results of X-ray and microstructure characterization were described in details in [4].

**2.2. Measurements.** For the investigations of polarization reversal (PR) and current-voltage characteristics, the measuring set for complete ferroelectric characterization [2] was used. The examination of PR characteristics was performed in two actual modes, namely, ferroelectric hysteresis loops of charge (Q-V-loops) and current (I-V-loops) were registered.

The measurements were performed by Sawyer-Tower like circuit [5] in the multicycle mode under applied a.c. triangular drive voltage  $V_d$  of 1 Hz of frequency in the amplitude range of  $0 \leq V_{da} \leq 11$  V.

To eliminate PR-current contribution, the examination of dynamic current-voltage characteristics (J-V-curves) was performed at unipolar saw-tooth drive voltage  $V_d$  with 0.5 s of durability and 1 Hz of repetitive frequency in the amplitude range of  $0 \leq V_{da} \leq 10$  V.

Drive voltage was applied to the circuits of series connection of tested sample with reference resistor for I-V-loops and J-V-curves and with reference capacitor for Q-V-loops. The corresponding characteristics were observed by means of two-beam digital storage oscilloscope YB 54060 operating in I-V-mode. The voltage on the reference elements was monitored and plotted versus drive voltage.

Piezoelectric response force microscopy (PFM) observations of deformation-voltage (X-V-) loops were carried out with a commercial AFM Veeco "Dimension 3100" using the probe of App-Nano ANSCM-PA type with cantilever spring constant of  $\approx 40$  N/m (resonance frequency  $\approx 300$  kHz). The probe was equipped by Si tip with double layer of chromium and platinum/iridium5 (Pt/Ir5) coating of 23 nm thick. After localization of the top Pt electrode by AFM imaging, the PFM measurements were performed in the contact mode with PFM tip located over the PZT grain ( $1-3 \mu\text{m}$ ) visible through the top Pt electrode.

A modulation ac voltage of 2 kHz of frequency and 1.5 V of amplitude was applied to the conductive PFM tip, and dc driving voltage  $V_d$  cycled in the range  $-10 \text{ V} \leq V_d \leq +10 \text{ V}$  was applied to the bottom Pt electrode of the film. The amplitude of  $V_d$  was chosen to be higher than the coercive voltage for polarization reversal in the examined PZT film.

PFM response amplitude hysteresis loops were recorded as a function of  $V_d$  by demodulating photoelectrically transformed cantilever deflection signal with a lock-in amplifier Signal Recovery 7270.

### 3. Results and Discussion

**3.1. Polarization Reversal Characteristics.** The obtained Q-V-loops and I-V-loops of polarization reversal are presented in Figures 1 and 2, respectively. Under increasing  $V_{da}$ , the start of saturation of Q-V-loops (Figure 1(a)) is accompanied by appearance of maxima on I-V-loops (Figure 2(a)). The main characteristics of Q-V- and I-V-loops are changed after several minutes of cycling under  $V_{da} = 10$  V that can be considered as forming.

**3.1.1. The Peculiarities of Bipolar Charge-Voltage Loops.** Under increasing  $V_{da}$ , the tendency of Q-V-loops for saturation is accompanied by vertical (along Q-axis) shift (Figure 1(a)), which reaches the maxima and starts to decrease under subsequent  $V_{da}$  increasing. At  $V_{da} = 10$  V, the coercive voltages  $V_c$  obtained from Q-V-loop are  $V_c^- = -3.4$  V and  $V_c^+ = +5.2$  V. The vertical shift changes its sign (see Figure 1(a)) under decreasing  $V_{da}$  after forming at  $V_{da} = 10$  V.

Figure 1(b) presents the  $V_{da}$  dependences of charge asymmetry factors  $\Delta Q_i = (Q_i^+ + Q_i^-)/2$  considered as a measure of the vertical shift of Q-V-loop ( $i = m, r$  correspond to maximal ( $V_d = V_{da}$ ) and remanent ( $V_d = 0$ ) polarization, resp.). Under  $V_{da}$  increasing the positive  $\Delta Q_m$  and  $\Delta Q_r$

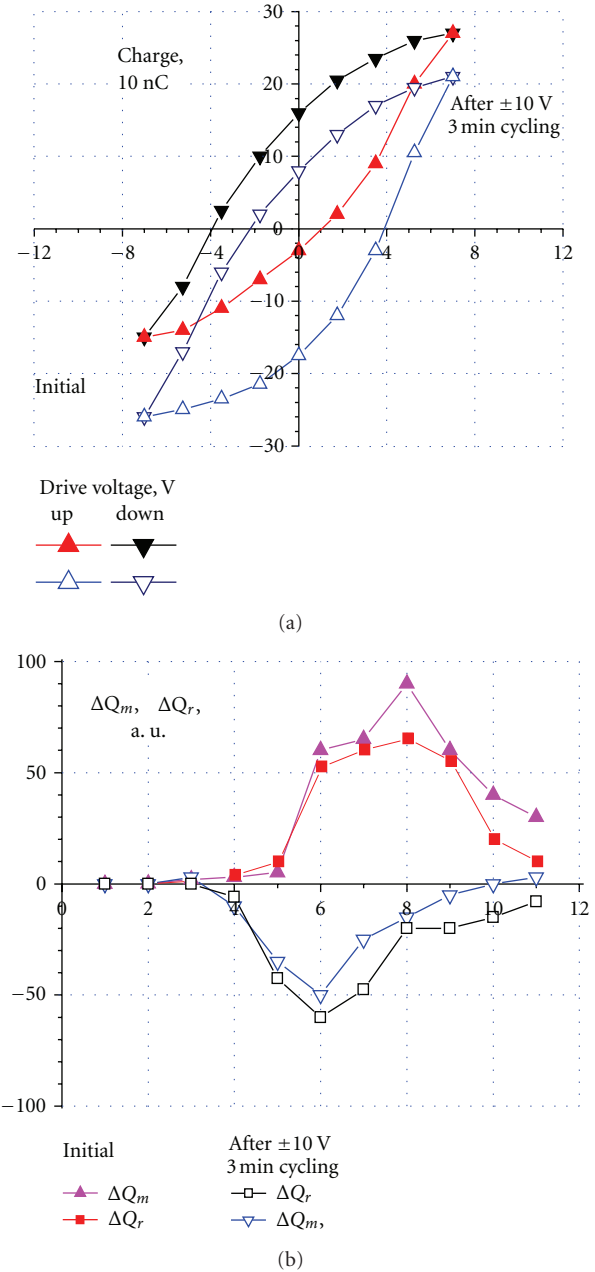


FIGURE 1: Bipolar charge-voltage loops of polarization reversal (a) and drive voltage dependences of charge asymmetry factors (b).

appear in  $V_c^+$  vicinity. After forming under subsequent  $V_{da}$  decreasing the negative  $\Delta Q_m$  and  $\Delta Q_r$  disappear in  $V_c^-$  vicinity. This behaviour of  $\Delta Q_m(V_{da})$  and  $\Delta Q_r(V_{da})$  shows that the vertical shift of Q-V-loops is connected with PR processes in the coercive  $V_c^-$  and  $V_c^+$  vicinities.

**3.1.2. The Peculiarities of Bipolar Current-Voltage Loops.** The observed I-V-loops are noticeably asymmetrical (Figure 2(a)). The absolute value of the voltage  $V_m^+$  of positive current maximum  $I_m^+$  is higher than that of the voltage  $V_m^-$  of the negative current maximum  $I_m^-$ . At that,  $I_m^+$  value is significantly

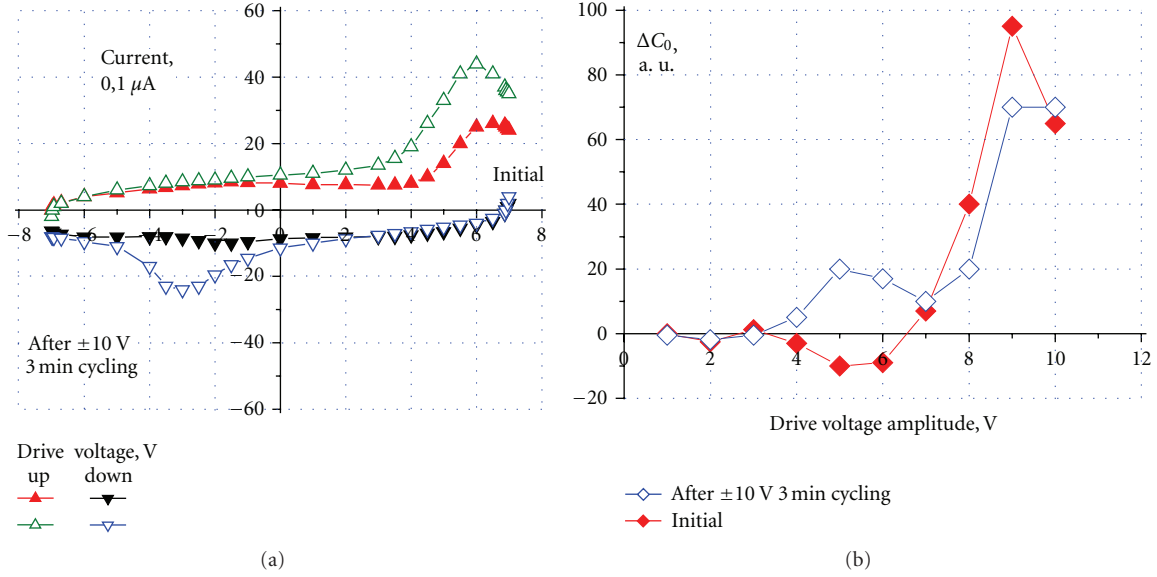


FIGURE 2: Bipolar current-voltage loops of polarization reversal (a) and drive voltage dependence of capacitive asymmetry factor (b).

higher than that of  $I_m^-$ . At  $V_{da} = 10$  V, the voltages  $V_m$  obtained from I-V-loop are  $V_m^- = -3.7$  V and  $V_m^+ = +6.7$  V.

The forming under  $V_{da} = 10$  V results in the change of  $V_m^+$  and  $V_m^-$  values and increase of  $I_m^+$  and  $I_m^-$  values under subsequent  $V_{da}$  decrease (see Figure 2(a)).

Because the vertical (along I-axis) size of I-V-loop is proportional to the sample capacity  $C$  [6], the value of  $\Delta I_0 = (I_0^- + I_0^+)/2$   $V_{da}$  (where  $I_0^-$  and  $I_0^+$  are the currents at  $V_d = 0$ ) can be considered as a measure of the capacitive asymmetry  $\Delta C_0 = (C_0^- - C_0^+)$  of I-V-loop.

Figure 2(b) presents  $V_{da}$  dependences of capacitive asymmetry factor  $\Delta C_0 \propto \Delta I_0$ . Under  $V_{da}$  increasing and its subsequent decreasing after forming the values of  $\Delta C_0$  are near the same at  $V_{da} < V_m^-$  and at  $V_{da} > V_m^+$ . However, in the range  $V_m^- < V_{da} < V_m^+$  not only the behaviour of  $\Delta C_0(V_{da})$  but also  $\Delta C_0$  signs are different before and after forming. At that, the maximal  $\Delta C_0$  difference before and after forming is observed at  $V_{da} \approx |V_c^+| = 5.2$  V. This behaviour of  $\Delta C_0(V_{da})$  shows that the current asymmetry of I-V-loops is connected with PR processes in  $V_m^-$  and  $V_m^+$  vicinities.

Direct modelling by means of the electrical circuit that includes inversely connected Zener diodes divided by parallel RC circuit [7] demonstrates the possibility of controlling the value of vertical shifts of Q-V-loops and peculiarities of I-V-loops by adjusting Zener voltages.

**3.2. Unipolar Current-Voltage Characteristics.** For the observed unipolar J-V-curves (Figure 3(a)) under  $V_{da}$  increasing and its subsequent decreasing after forming is characteristic of the change of asymmetry degree and the shape of the positive and negative branches and also the change of the voltages  $V_t^+$  and  $V_t^-$  of transition between sublinear and superlinear J-V-regions.

As a measure of J-V-curve asymmetry the ratio  $A_R = J_m^+/J_m^-$  of the maximal (index “m”) current values of positive  $J_m^+(V_{da})$  and negative  $J_m^-(V_{da})$  branches was taken.

The  $V_{da}$  dependences of  $A_R$  and also  $V_t^-/V_{da}$  and  $V_t^+/V_{da}$  for initial and formed J-V-curves are presented in Figure 3(b). The values of  $A_R$  for initial and formed J-V-curves are different at  $V_{da} < V_m^+$  and became near equal at  $V_{da} \geq V_m^+$ . Under increasing  $V_{da}$  the dependence  $A_R(V_{da})$  reaches the maximum at  $V_{da} \approx |V_c^-|$ . After forming  $A_R(V_{da})$  dependence has the smooth step with the start at  $V_m^-$ , the centre near  $V_{da} \approx V_c^+$ , and the finish at  $V_m^+$ .

The difference in  $V_t^+(V_{da})$  and  $V_t^-(V_{da})$  behaviour is displayed by  $V_{da}$  dependences of  $V_t^+/V_{da}$  and  $V_t^-/V_{da}$  ratios (see Figure 3(b)). The equality of  $V_t^+/V_{da}$  and  $V_t^-/V_{da}$  is observed up to  $V_{dm} \approx |V_c^-|$  and the value of  $(V_t^+ - V_t^-)/V_{da}$  reaches maximum at  $V_{da} \approx V_m^+$ .

This behaviour of asymmetry ratio  $A_R(V_{da})$  and also transition voltages  $V_t^+(V_{da})$  and  $V_t^-(V_{da})$  of unipolar J-V-curves manifests the peculiarities in the coercive  $V_c^-$  and  $V_c^+$  vicinities as bipolar Q-V-loops and in  $V_m^+$  and  $V_m^-$  vicinities as bipolar I-V-loops.

**3.3. Deformation-Voltage Loops.** Figure 4 presents the local deformation-voltage (X-V-) loops (PFM-amplitude) studied on the same Pt/PZT/Pt:Ti/SiO<sub>2</sub>/Si-substrate structure as were examined integral charge Q-V- and current I-V-loops of polarization reversal.

As it is seen from Figure 4(a), the initial X-V-loop is remarkably asymmetrical. An apparent approach of the loop shape to a near symmetrical butterfly-like one (Figure 4(b)) was achieved by cycling under  $V_d$  sweeping with a decreasing from 10 V to zero amplitude (local forming procedure).

So, the forming procedure results in similar symmetrization of corresponding characteristics related to integral and local PR processes.

## 4. Conclusion

The developed conception of asymmetry factors clearly shows the interconnection of polarization reversal asymmetry and

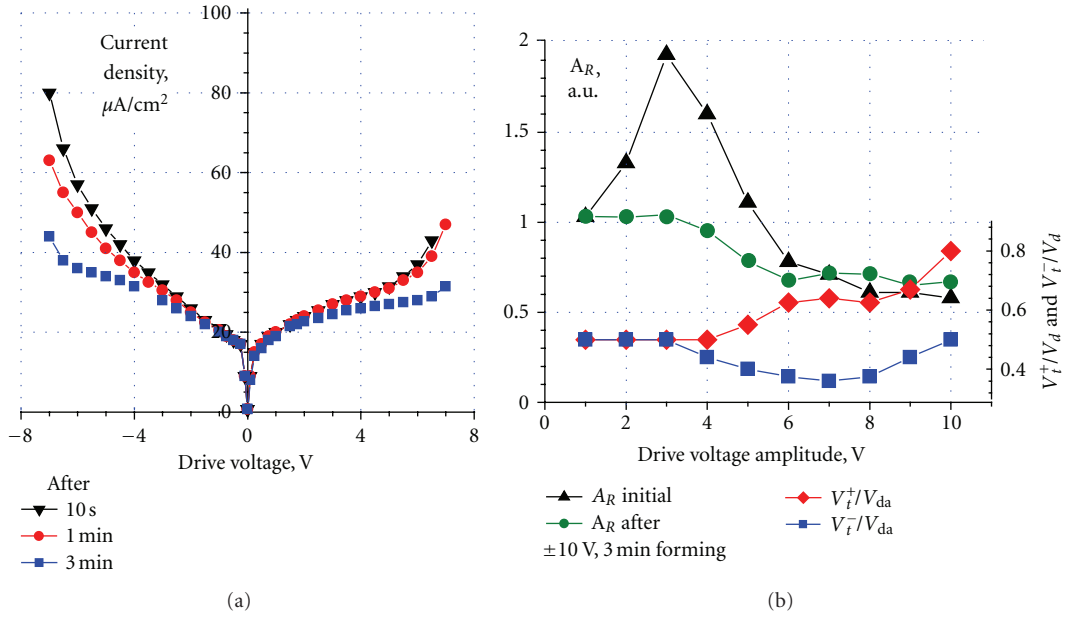


FIGURE 3: Unipolar current-voltage curves (a) and drive voltage dependences of transition voltages and asymmetry ratio (b).

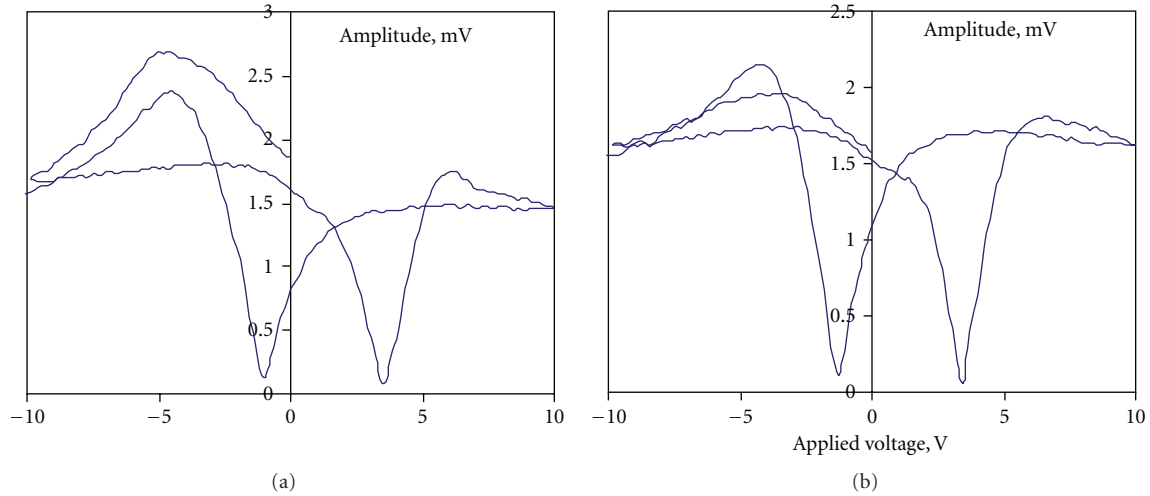


FIGURE 4: PFM amplitude-voltage loops initial (a) and after forming by drive voltage sweeping with decreasing amplitude (b).

asymmetry of electrical space charge transfer in ferroelectric Pt/PZT-film/Pt:Ti/SiO<sub>2</sub>/Si-substrate structures.

The obtained results evidence the interrelation of charge transfer and polarization reversal processes in PZT-film on Si structures. It is assumed that charge transfer-polarization reversal interplay takes place at least in the case of polarization reversal assisted by charged domain wall displacement proper to PZT films [8].

The physical reason of the observed consequences of the electrical treatment is the space charge transfer symmetrization through the film thickness under alternative voltage forming.

The observed asymmetry of the investigated polarization reversal and current-voltage characteristics is developed in the conditions of the space charge transfer in time.

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