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Dielectric characteristics of novel composite nanomaterial

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Characteristic features of the $PbZr_{0.54}Ti_{0.46}O_3$ formation in etched swift heavy ion tracks and investigations of dielectric properties of the obtained structures are reported in this work. $PbZr_{0.54}Ti_{0.46}O_3$ compound is formed as a result of thermal treatment of film structures with identical composition being ion-beam sputtered on Si/SiO_2 substrates. During the temperature dependence studies of the Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure at various frequencies their dispersion is obtained. It is shown that in the temperature range from 360 to 406 °C the frequency dependence of the dielectric permittivity reaches maximum values which is explained by the presence of a ferroelectric phase transition.

1. Introduction

Recently, non-traditional methods of nanostuctures and their arrays formation are undergoing an intensive development. One of such methods, which was developed rather recently, being very prospective, is the swift heavy ion (SHI) tracks technology. It is based on the formation of narrow extended regions of radiation damage („latent ion tracks") in various dielectric materials under the influence of the SHI [1] and the eventual subsequent removal of the radiation-damaged zones by etching, to form narrow parallel extended nanopores in the corresponding dielectric material. During the complex investigations of Si / SiO_2 structures with embedded SHI tracks a new family of nanoelectronic systems was formed which has got a conditional name "TEMPOS" ("Tunable Electronic Material in Pores in Oxide on Semiconductors") [2 – 5].

A careful analysis of recent publications indicates the possibility to create nanosensor arrays on the base of "TEMPOS" systems, containing lead zirconate-titanate $PbZr_{0.54}Ti_{0.46}O_3$ (PZT) in etched SHI tracks. This can be explained by a number of functional electronic devices, such as ferro- and pyroelectric detectors, infrared detector cells, volatile memory devices, dynamic random-access memory devices, high-capacitance condensers, etc. [6 – 13], where this material can be used. However, for a successful application of such nanostructures it is necessary to investigate their dielectric properties as well. In frame of this work temperature and frequency dependences of

dielectric permittivity and dielectric loss tangent of Si/SiO$_2$ (PbZr$_{0.54}$Ti$_{0.46}$O$_3$) have been investigated.

2. Experiment

Single-crystal silicon "KEF-4.5" wafers with (100) orientation were used as initial substrates for the formation of experimental samples. Prior to the thermal treatment, the wafers were processed in acidic – peroxide mixture during 1 min, and then the treatment continued in peroxide-ammonium mixture during 10 min. Silicon oxide layer with thickness of 0.7±0.1 μm was formed by a thermal oxidation of Si under 1100 °C and oxygen pressure pO$_2$ = 10^5 Pa during 10 h.

For the formation of SHI tracks the Si(100)/SiO$_2$ structure was irradiated by ^{197}Au^{26+} ions with energy of 350 MeV and a fluence of 5×10^8 cm^{-2} at the Ion Beam Laboratory ("ISL") of the Helmholtz Centre Berlin for Materials and Energy (Germany). Ion tracks (ITs), which were formed in the SiO$_2$ layers as a result of irradiation, have been etched in fluoric acid with a concentration 1.35 wt.% at 20°C during 40 min. Due to the difference in etching rates of irradiated and non-irradiated areas of silicon oxide, stochastically distributed pores in the form of frustums with an average cross-section of 100 nm have been created there (Fig. 1).

The sputtering of PbZr$_{0.54}$Ti$_{0.46}$O$_3$ films on Si(100)/SiO$_2$ structure was carried out by the ion-beam method on the vacuum setup "Z 400" by "Leybold-Heraeus", which was equipped with an oil-free pumping system with turbomolecular pumps. Ceramic discs having composition PbZr$_{0.54}$Ti$_{0.46}$O$_3$+0,1PbO, diameter 80 mm and thickness 10 mm, served as targets. Two independent ion beams were used for the sputtering of films. The first beam was used for a preliminary cleaning of the surface, and the second beam was used for the sputtering of the target. A mixture Ar:O$_2$ prepared in the ratio 1:1 at the residual pressure in the chamber 10^{-3} Pa was used as a working gas. Magnetic field was formed by a solenoid of copper wire with a diameter of 0.8 mm in a fluoroplastic insulation wound on a copper core. Cleaning of the surface has been carried out by Ar$^+$ ions with energies of 800 – 1200 eV during 15–20 minutes at the discharge current 80 mA. For the compensation of positive charge on the target surface, a tungsten wire with diameter 0.5 mm was placed near the target. Current values there were about 12 – 16 A. Sputtering of the target has been carried out by ions of argon and oxygen with energies of 1800 - 2400 eV at current 30 mA.

Chemical deposition of metals Pb, Zr and Ti from a solution in the ITs of SiO_2 layer was carried out at room temperature. With the goal of an increase of efficiency of this process the samples underwent sensibilization and activation procedures, i.e. the formation of corresponding nucleation centers on IT walls has been provided. $[Pd(NH_3)_4]^{2+}$ (with Cl^- counter-ion), sensitizing complex was used, which being limited by a corresponding agent, "bound" Pd atom to silicon oxide by means of complex formation with surface amines, carbonyls and hydroxides, forming such groups as COOPd [14, 15]. Annealing of obtained samples in oxidizing medium has been carried out for the formation of $PbZr_{0.54}Ti_{0.46}O_3$ compound. Temperature in heating set ups has been maintained by "RIF-101" system and controlled by Pt–Pt/Rh(10%) thermocouple with the accuracy ± 0.5 K. From the structures prepared in this way samples having rectangular form with length 20 mm and width 10 mm have been cut.

Gold electrical contacts were deposited on the investigated samples surface from the side of SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) using thermal evaporation (substrate temperature 300°C). The thus prepared "TEMPOS" structures exhibit a multitude of current paths as well in horizontal (between both surface contacts across the deposited PZT layer and in the semiconducting channel below the SiO_2/Si interface) as in vertical direction (between the surface PZT layer and the Si substrate across the parallel PZT-filled etched ion tracks, the PZT/Si interface and the Si substrate), Fig. 2. Upon application of suitable potentials at both the surface electrodes and the Si backing, different competing currents emerge across the individual current paths, which may lead to peculiar collective electronic effects of the given device, such as rectification, negative differential resistances, periodic or random charge spike emission and others. Embedded in adequate electronic circuits, this can be used to create amplifiers, periodic or random pulse generators, sensors, nanometric point light sources, frequency filters, logic AND/OR switches, etc. [2-5].

The phase composition of $PbZr_{0.54}Ti_{0.46}O_3$ films has been studied by X-ray phase analysis with the "Philips X' Pert" setup in Cu K_α irradiation, with the use of the "ICSD-PDF2" ("Release 2000") database. The microstructure of the samples with $PbZr_{0.54}Ti_{0.46}O_3$ in ITs of SiO_2 on silicon substrate during various stages of their preparation was investigated by scanning electron microscopy (SEM) by means of the "JEOL JSM 6360" set up.

For the control of the structure formation process of $PbZr_{0.54}Ti_{0.46}O_3$ in ITs X-ray photoelectron spectroscopy (XPS) measurements were carried out using the "Physical

Electronics ESCA 5700" setup with a non-monochromated Al K$_\alpha$ X-ray source. The analysis area was ca. 0.5 mm^2. The upper contaminating layer was removed with Ar$^+$ ion bombardment at 4 kV acceleration voltage. Calibrations with different metal oxides revealed removal rates of about 1.2 nm/min. The possible emerging sample charging which would affect the emission of photoelectrons was compensated by shifting the spectra to Ti 2p3/2 at 458.2 eV (binding energy of PbTiO$_3$).

Local ferroelectric properties of the samples were investigated with piezoresponse force microscopy (PFM) using a commercial "NTEGRA Aura" (NT-MDT) setup equipped with an external lock-in amplifier ("SR-830A", Stanford Research) and a function generator ("FG-120", Yokogawa). A commercial silicon cantilever ("Nanosensor PPP-NCHR") with the spring constant k=42 N/m and with tip radius less than 10 nm was used. The measurements were performed under an applied AC voltage with the amplitude $V_{ac} = 2.5$ V and frequency f = 50 kHz.

In PFM experiments, an AC electric field is applied between a bottom electrode and a conducting tip used as a moveable electrode. During the measurements a probing tip is in mechanical contact with the sample and follows the piezoelectric motion of the sample surface. Thus, the amplitude of the tip vibration measured by the lock-in amplifier provided information on the piezoelectric strain.

For investigations of the dielectric permittivity (ε) and the dielectric loss tangent (tgδ) Si/SiO$_2$ (PbZr$_{0.54}$Ti$_{0.46}$O$_3$) samples were placed in a cell (schematically given in Fig.2), where the measurements were carried out. Contact needles in temperature range 50 – 500 °C and frequency range $5\times10^3 – 8\times10^5$ Hz were used in the "Hewlett-Packard – 4192 A" set up, containing thermometer "Keithley-740" and a temperature controller "Tabai STPH-100".

3. Results and Discussion

Optimization of conditions of synthesis of PbZr$_{0.54}$Ti$_{0.46}$O$_3$ compound in ion tracks after chemical deposition of metals Pb, Zr and Ti was carried out on film structures with identical composition, sputtered by the ion-beam method on Si/SiO$_2$ substrates.

PbZr$_{0.54}$Ti$_{0.46}$O$_3$ films sputtered with a rate of 6 – 9 nm / min on a cold substrate (T$_{substr}$ ~ 30 °C) of oxidized silicon had a quasiamorphous structure (Fig. 3). Besides, as at the vacuum sputtering method film growth was carried out at low values of pO$_2$ ~ 10^{-5} Pa, a depletion of oxygen in the films composition took place. Therefore, in order

to saturate the films with oxygen to acquire the $PbZr_{0.54}Ti_{0.46}O_3$ ferroelectric phase, as well as for a decrease of the quantity of re-evaporated lead from their surface, thermal annealing processes (450 – 600º C) were carried out at increased oxygen pressure ($pO_2 = 2 \times 10^5$ Pa during 30 min). Additionally, the lead oxide was introduced in the target composition. This was made in such a way that a decrease of the amount of lead atoms below from the stoichiometric composition of 2 mole % would render the formation of the perovskite phase impossible. From the other hand, according to the X-ray phase analysis the increase of the PbO concentration above 10 mole % in $PbZr_{0.54}Ti_{0.46}O_3$ targets is not desirable because it would promote Pb segregation on grain boundaries of the target. This would change the cation relation and correspondingly violate of the required composition of the sputtered dielectric layers. The initial structure formation of $PbZr_{0.54}Ti_{0.46}O_3$ takes place at annealing with $T_{anneal} > 450°$ C and $pO_2 = 2\times10^5$ Pa. Nevertheless, under the given annealing conditions the films are not structurally homogeneous, which requires further increase of the annealing temperature. Most films were therefore annealed in the temperature region $500 < T_{anneal} \leq 550º$ C. A further increase of T_{anneal} leads to a decrease of the Pb content in the films.

The magnetron sputtering method allowed us to get homogeneously deposited films with excellently smooth sample surfaces. Atomic force microscopy (AFM) measurements performed in semicontact mode confirmed the small roughness of the sample surface (Fig. 4 a). The estimated RMS (root mean square) height of the surface is less than 20 nm whereas the average calculated grain size is about 100 - 300 nm.

The homogeneous deposition of the film has been tested by the AFM measurements performed in different parts of the sample thus confirming the plausibility of the film formation by the selected sintering method. Piezoresponse force microscopy measurements did not reveal essential polarization of the sample.

The amplitude of the piezoresponse signal is quite low and noisy (not shown), while it is enough to distinguish distinct ferroelectric domains with different polarization. Nevertheless a certain sample polarization was confirmed by piezoelectric loop measurements (Fig. 4 b). Strain versus DC voltage curves ($d_{33}(E)$) were measured in the so-called "pulse DC mode". Along with a linear component of the $d_{33}(E)$ loops affirming paraelectricity the obtained data also testify a spontaneous polarization of the sample. Based on the piezoelectric hysteresis loops we can argue about some remnant polarization of the sample [11]. The obtained ferroelectric response of the sample is smaller than the PFM signal observed for commercial PZT (52/48) films by orders of magnitude.

For the formation of $PbZr_{0.54}Ti_{0.46}O_3$ compounds the ITs of the SiO_2 layer the same thermal treatment regimes as those for $PbZr_{0.54}Ti_{0.46}O_3$ films have been used. The control of the evolution of $PbZr_{0.54}Ti_{0.46}O_3$ structures formation in ITs after the annealing processes of various duration was carried out by the XPS. This method makes it possible to estimate the kinetic energy of inner electrons or valence electrons, knocked out by a quantum of electromagnetic irradiation with known energy [16, 17]. When studying of the XPS spectra concerning the Ti – O bonds with energy ~ 458 eV in $PbZr_{0.54}Ti_{0.46}O_3$ compound a development of the structure formation was observed (Fig. 5). The largest maximum of the XPS spectra relates to the sample annealing for 8 min. Corresponding maxima on the XPS spectra were observed for Zr at a binding energy ~ 182 eV and for Pb at a binding energy ~ 138 eV (Fig. 6 b, c). One can suppose that during the $PbZr_{0.54}Ti_{0.46}O_3$ structure formation the deficient oxygen is absorbed from the gaseous phase and it arrives at the structure by means of surface diffusion along the ITs walls (which have a potential energy below that of the planar surface).

During the measurements of the temperature dependence of the dielectric permittivity of the Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure in the temperature range T = 0 – 500°C at various frequencies ($5\times10^3 – 8\times10^5$ Hz) it was found that ε values increase weakly in the temperature range 100 - 250°C and in the low-frequency area ($\nu=5\times10^3$ Hz and $\nu = 4\times10^4$ Hz), Fig. 7. At T > 250°C a sharp jump of ε up to the values of $\varepsilon_{max} = 8.17\times10^3$ (at T=375° C and $\nu=5\times10^3$ Hz), $\varepsilon_{max} =9.3\times10^3$ (at T = 380°C and $\nu = 4\times10^5$ Hz) and $\varepsilon_{max} =10.1\times10^3$ (T=372° C, $\nu=8\times10^5$ Hz) takes place. It should be noted that these maxima reveal the tendency of a shift toward increasing temperature with a decrease of the measurement frequency. One could suppose that the appearance of dielectric permittivity maxima in the temperature range 370 - 380°C is related to a ferroelectric phase transition (FPT) in $PbZr_{0.54}Ti_{0.46}O_3$ compounds, which leads to disorder in the cation sublattice [6, 8].

The frequency dependence of the dielectric permittivity shows the existence of a dispersion $\varepsilon(\nu)$, which is especially pronounced in that temperature range where the maximum ε values, where ε_{max} values decrease with frequency rise. Such a dependence indicates the existence of relaxation polarization at low frequencies in the Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure. This effect makes a contribution to the static dielectric permittivity, preconditioning losses at low frequencies.

In that case every passage of a cation from a site to a void creates a dipole moment. If the number of voids in a crystal lattice is large, the dipole can have an arbitrary direction and change it in the external electric field creating the relaxation polarization. Without any interaction between dipoles the disordering effect would not lead to any occurrence of the FPT [11]. However, with increasing dipole concentration the interdipole interaction lowers the energy of dipole formation, so that FPT is observed, and therefore this effect should also take place in the Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure.

The data of the frequency dependence of the dielectric loss tangent, which are given in Fig. 8, show a quasi-oscillating nature. Beginning with 100°C, the $tg\delta(T)$ dependences for all the measured frequencies in the range ($5\times10^3 - 8\times10^5$ Hz) undergoes a monotonic increase, achieving several small maxima around 200°C. Their appearance is possibly related to the ferroelectric properties of $PbZr_{0.54}Ti_{0.46}O_3$ due to the absence of composition inhomogenities there. Similarly to the $\varepsilon(T)$ dependence at various frequencies, all the $tg\delta(T)$ maxima show the tendency to a shift towards increasing temperature. This tendency is especially pronounced for the second maximum and observed for all frequencies. In that case the largest maximum of $tg\delta(T)$ for the measured frequency 5×10^3 Hz is located at the temperature ~ 330°C. The further monotonic increase of the $tg\delta(T)$ dependence indicates an even larger third maximum for all the measured frequencies. Such a behaviour of $tg\delta(T)$ obviously confirms the presence of the FPT, caused by a disorder of the cation sublattice in the Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure as well as in the case of $\varepsilon(T)$ investigations.

4. Conclusions

Following conclusions can be done in frames of this work:

– the structure of $PbZr_{0.54}Ti_{0.46}O_3$ films, obtained by ion-beam sputtering on Si/SiO_2 substrates with rates of 6 - 9 nm/min and a substrate temperature of ~ 30 °C is quasiamorphous. Structure formation in the films takes place at annealing temperatures $T_{anneal} > 450$ °C and an oxygen pressure $pO_2=2\times10^5$ Pa. The films obtained in the temperature range $500 < T_{anneal} \leq 550$ ºC have the best homogeneity;

– chemical deposition of Pb, Zr and Ti metals within etched swift heavy ion tracks in SiO_2 thin layers on silicon substrates with a subsequent annealing at $T_{anneal} = 550$ °C and $pO_2 = 2 \times10^5$ Pa make it possible to obtain $PbZr_{0.54}Ti_{0.46}O_3$ compounds therein;

- during investigations of dielectric properties of the Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure at various frequencies (5×10^3 Hz, 4×10^4 Hz and 8×10^5 Hz) a dispersion of the dielectric permittivity is revealed. The $\varepsilon=f(\nu,T)$ dependence shows maxima in the temperature range 360 – 406 ºC ;

- it is shown that maximum values of the dielectric permittivity at T = 360 – 406 ºC at various measured frequencies are caused by the presence of a ferroelectric phase transition in $PbZr_{0.54}Ti_{0.46}O_3$ compound, which leads to a disorder in the cation sublattice and the disappearance of ferroelectric properties of the structure. Temperature dependences of the dielectric loss tangent obtained at various frequencies indicate the presence of a ferroelectric phase transition as well;

- suitable optimized Si/SiO_2(PZT) structures of the "TEMPOS" type can be prepared, which should enable applications in quite a number of advanced functional electronic devices, as mentioned above.

Funding

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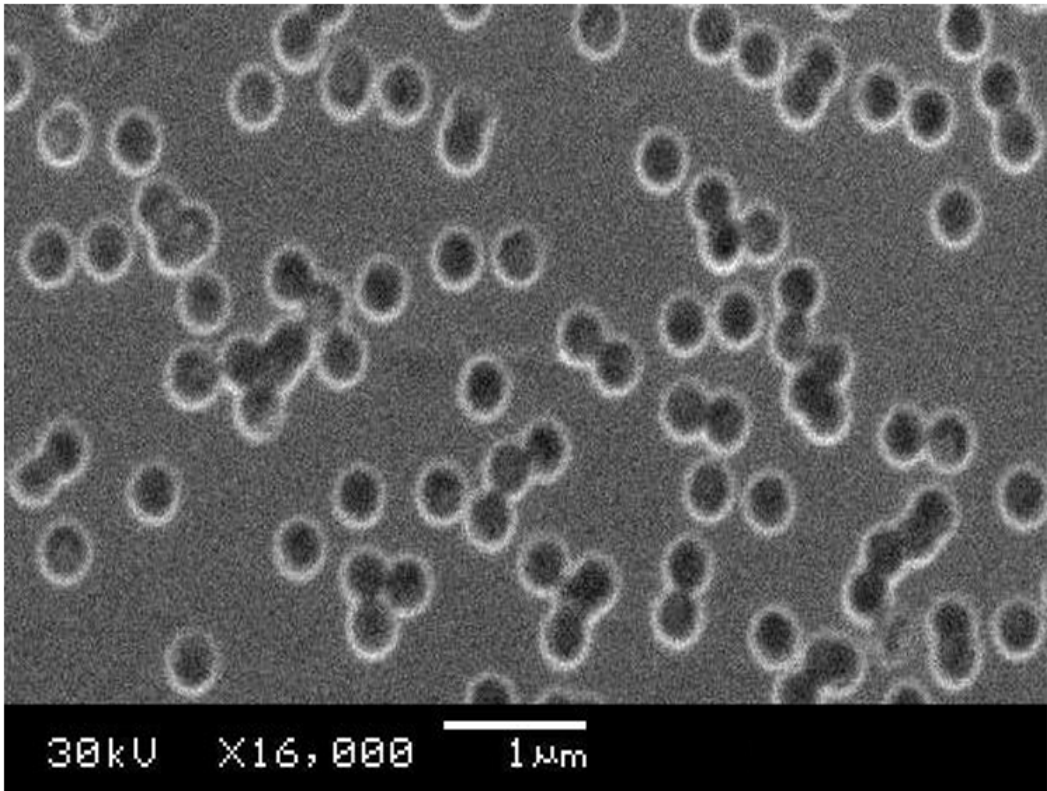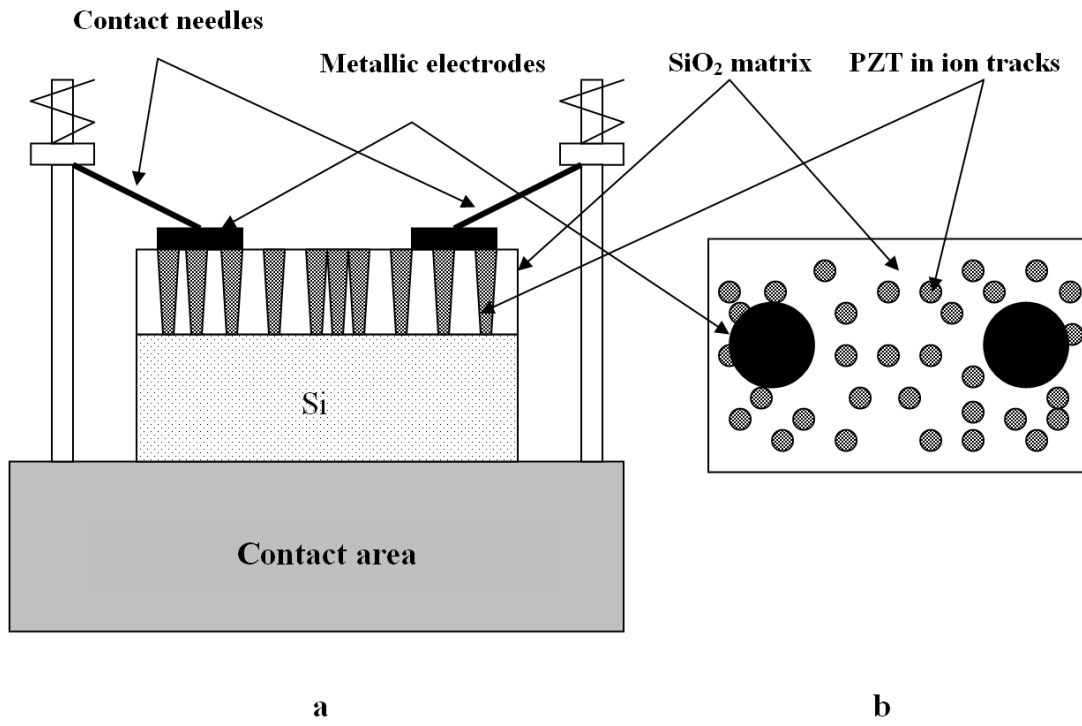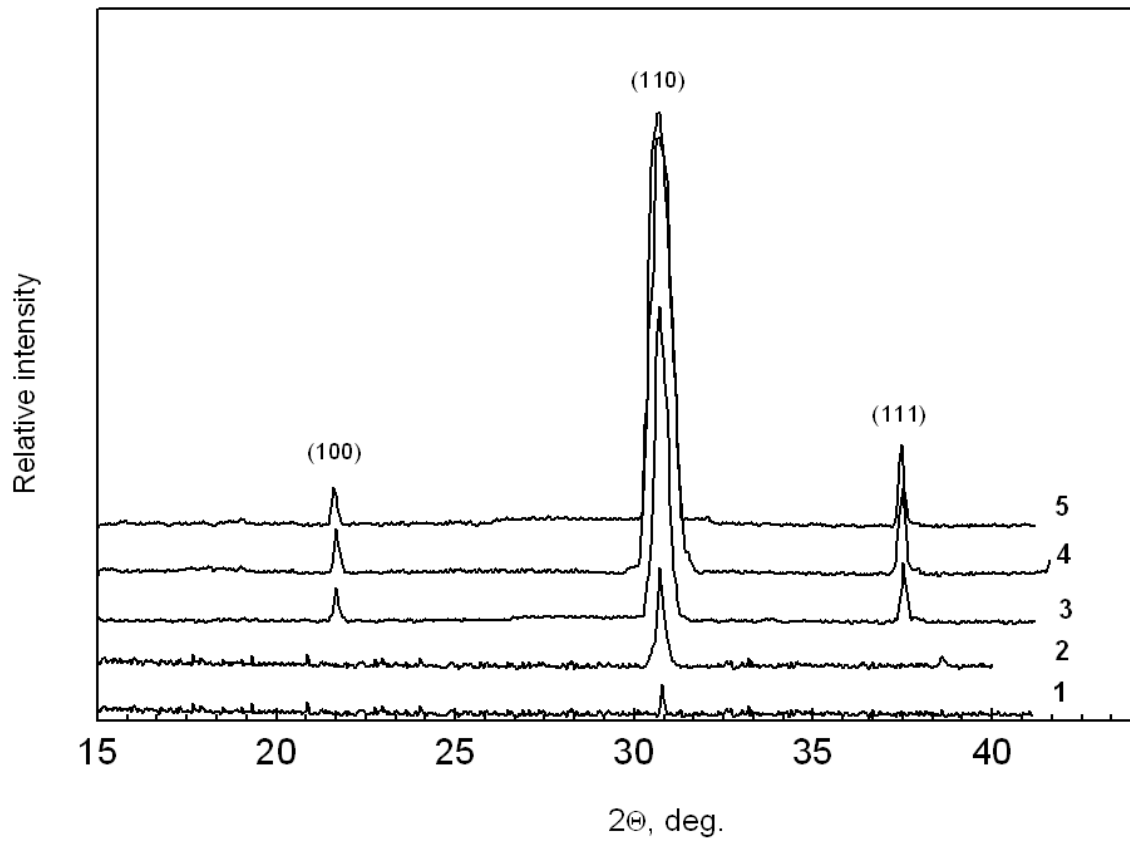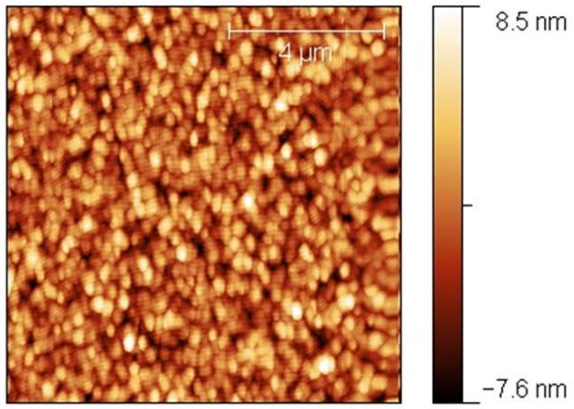
Figures

Figure 1

Contact needles

Metallic electrodes

SiO$_2$ matrix

PZT in ion tracks

Si

Contact area

a

b
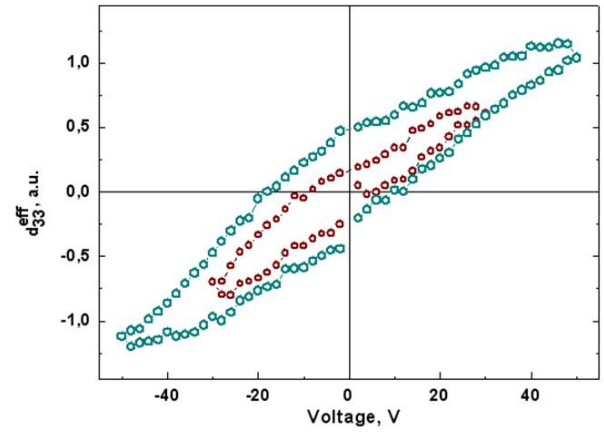
Figure 2

Figure 3

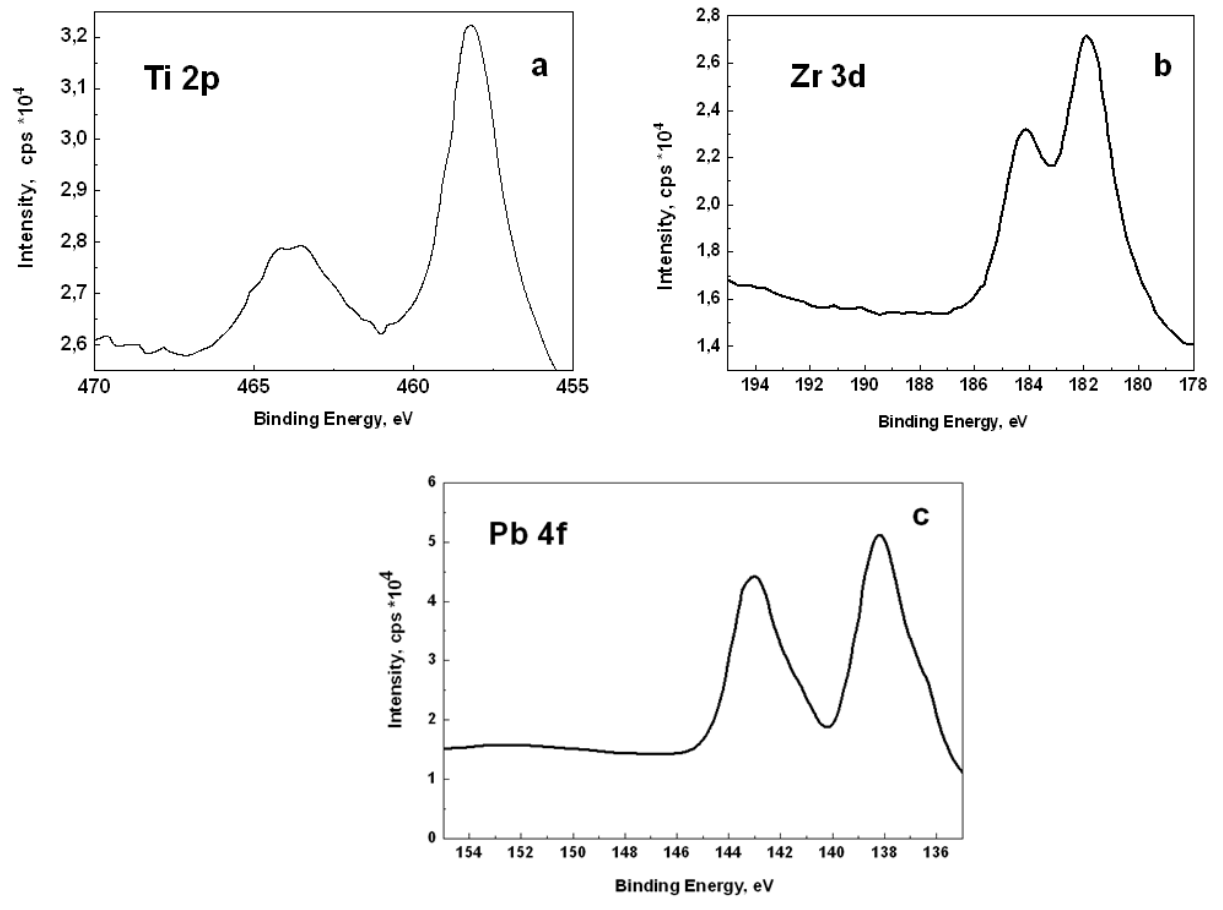| a | b |

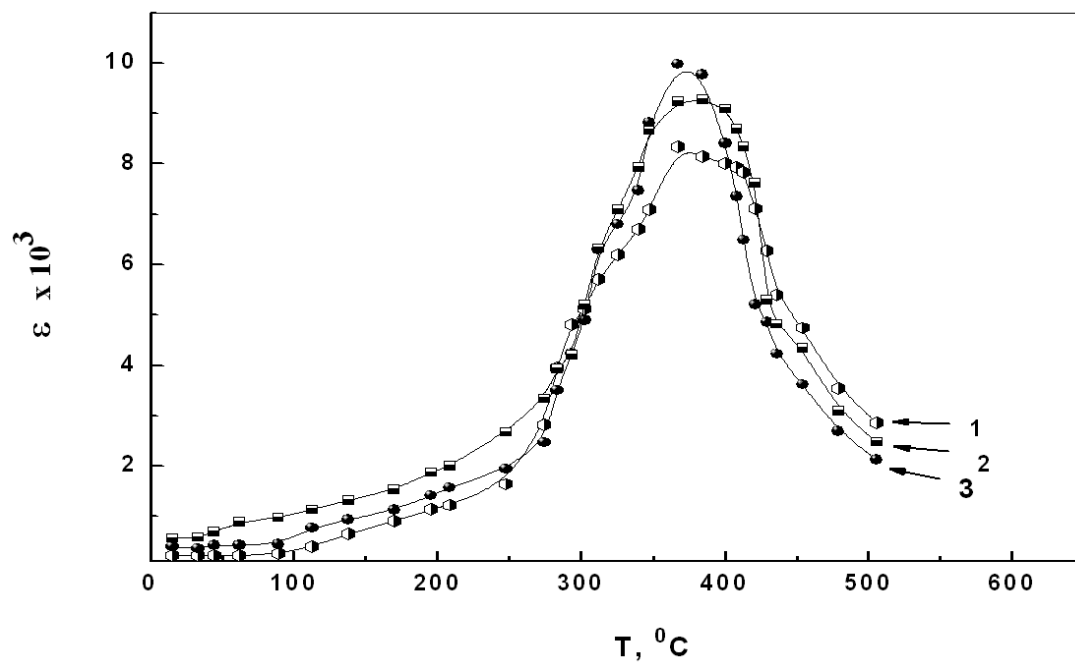**Figure 4**
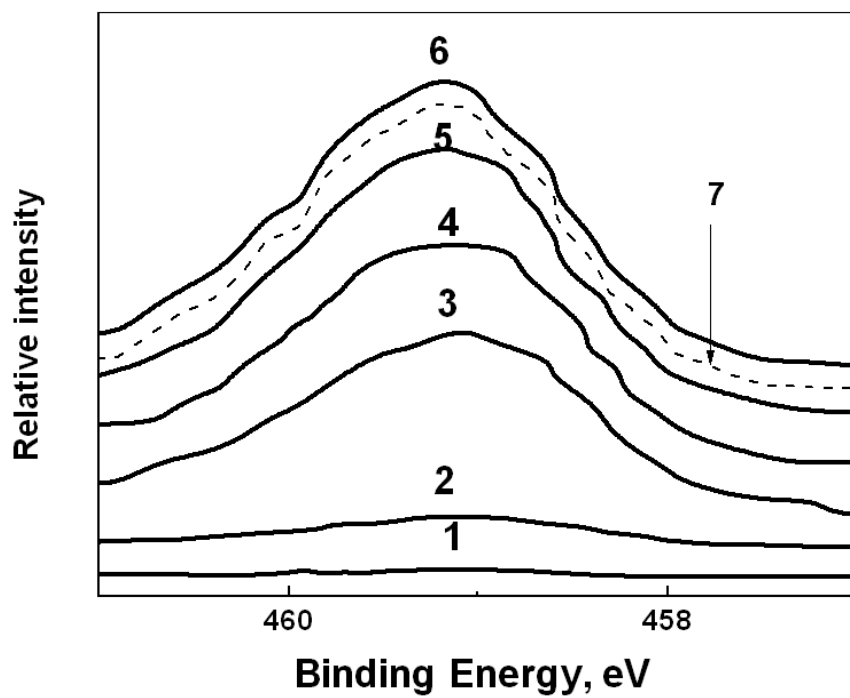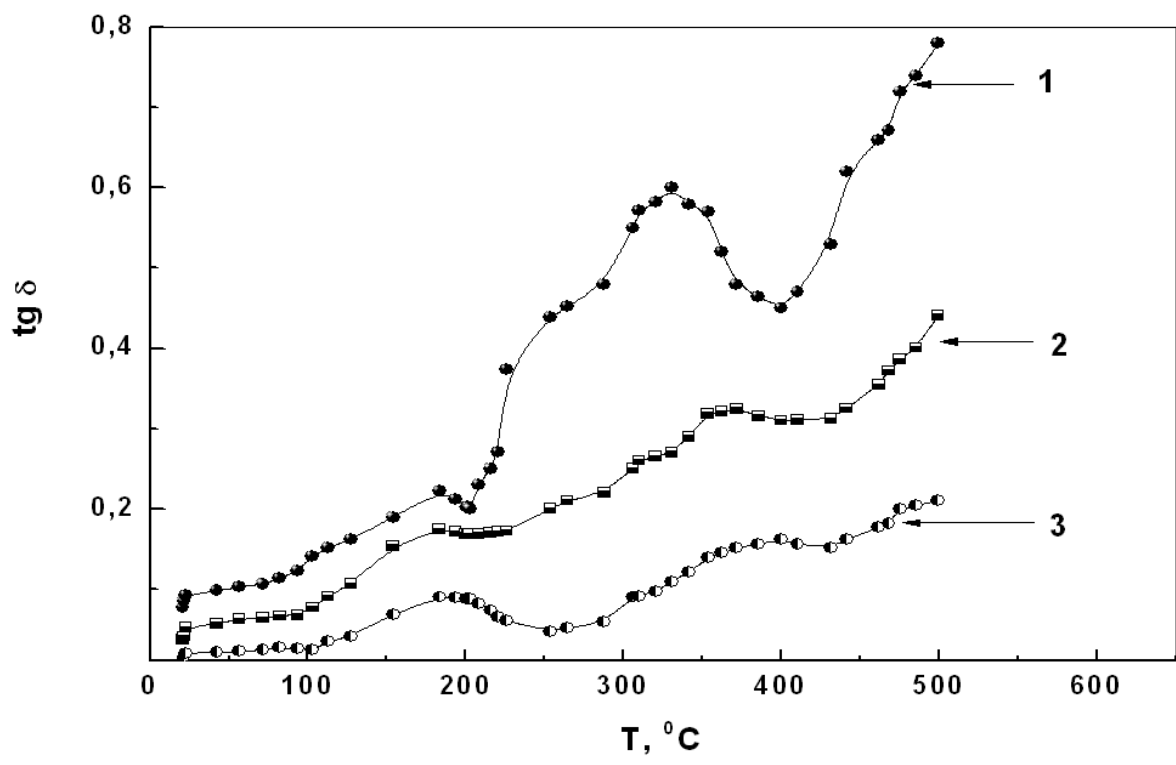
Figure 5

Figure 6

Figure 7

Figure 8

Figure Captions

Figure 1. A surface of the sample with ion tracks in SiO_2 layer. Scanning electron microscopy   image.

Figure 2. A diagram of the measuring cell of the "TEMPOS" system on the base of Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure : (a) side view, (b) top view

Figure 3. XRD spectra of $PbZr_{0.54}Ti_{0.46}O_3$ films surface obtained at various annealing temperatures: without annealing (1), 450 °C (2), 500 °C (3), 550 °C (4), 600 °C

Figure 4. AFM image (a) and piezoresponce-force microscopy loops (b) for the multilayered structure $Si/SiO_2/Ti/Pt/$ $PbZr_{0.54}Ti_{0.46}O_3/Pt$ annealed at 550 °C during 0.5 h in O_2

Figure 5. The change of XPS spectra maximum values in $PbZr_{0.54}Ti_{0.46}O_3$ at $T_{anneal}=550$°C and $pO_2=2\times10^5$ Pa, with various duration of annealing times: 3 min (1), 4 min (2), 5 min (3), 6 min (4), 7 min (5), 8 min (6), 9 min (7)

Figure 6. XPS results for $PbZr_{0.54}Ti_{0.46}O_3$ of a $Si/SiO_2(PbZr_{0.54}Ti_{0.46}O_3)$ structure at $T_{anneal}=550$°C during 8 min for Ti (a), Zr (b) and Pb (c)

Figure 7. Temperature dependences of the dielectric permittivity of Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure at frequencies: 5×10^3 Hz (1), 4×10^4 Hz (2) and 8×10^5 Hz (3)

Figure 8. Temperature dependences of the dielectric loss tangent of Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure at frequencies: 5×10^3 Hz (1), 4×10^4 Hz (2) and 8×10^5 Hz (3)