DIELECTRIC PROPERTIES OF $PbZr_{0.54}Ti_{0.46}O_3$ IN SWIFT HEAVY ION TRACKS OF Si/SiO_2 NANOSTRUCTURES

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Characteristic features of formation of $PbZr_{0.54}Ti_{0.46}O_3$ compound in etched swift heavy ion tracks obtained by irradiation of Si/SiO_2 structure by $^{197}Au^{26+}$ ions and investigations of dielectric properties of the obtained structures are considered. $PbZr_{0.54}Ti_{0.46}O_3$ compound is obtained as a result of optimization of thermal treatment of film structures with identical to the sputter target composition, being ion-beam sputtered on Si/SiO_2 substrates. During the study of temperature dependence of Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure at various frequencies (5×10^3 Hz – 8×10^5 Hz) its dispersion is obtained.

1. Introduction

At present non-traditional methods of creation of nanostuctures and their arrays are undergoing intensive development. One of such methods, which elaboration was started rather recently and being very prospective is the swift heavy ion (SHI) tracks technology [1]. In the process of complex investigations of Si/SiO_2 structures with the 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) [2,3]. Analysis of the publications released in the last years confirms the prospect of a creation of 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, for their use in a number of functional electronic devices, such as ferro- and pyroelectric detectors, infrared detector cells, volatile memory devices, *etc*. [4,5]. Nevertheless, for a successful employment of nanostructures on the base of Si/SiO_2 and PZT it is necessary to investigate their dielectric properties. This was a goal of a research reported in the present work.

2. Experimental

The sputtering of $PbZr_{0.54}Ti_{0.46}O_3$ films on Si(100)/SiO_2 structure was carried out by ion-beam method on the vacuum setup “Z 400” by “Leybold-Heraeus”. Ceramic discs having the composition $PbZr_{0.54}Ti_{0.46}O_3$+0.1PbO, the diameter of 80 mm and the thickness of 10 mm served as targets. Sputtering of the target has been carried out by argon and oxygen ions with the energies of 1800-2400 eV at the current of 30 mA.

For the formation of SHI tracks the Si(100)/SiO_2 structure was irradiated by $^{197}Au^{26+}$ ions with the energy of 350 MeV and the fluence of 5×10^8 cm^{-2} at the Helmholtz Centre Berlin for Materials and Energy (Germany). Ion tracks (ITs) which were formed in SiO_2 layer as a result of irradiation were etched in the fluoric acid with the concentration of 1.35 mass.% at 20 °C during 40 min. Due to a 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. Thereafter a chemical deposition of metals Pb, Zr and Ti from a solution in the ITs of SiO_2 layer was carried out. Then, an oxygen annealing of the samples was realized for the formation of $PbZr_{0.54}Ti_{0.46}O_3$ compound.

For the monitoring of structure formation process of $PbZr_{0.54}Ti_{0.46}O_3$ in ITs the X-ray photoelectron spectroscopy (XPS) measurements were carried out by means of the “Physical Electronics ESCA 5700” setup with a non-monochromated Al K_α X-ray source. Measurements of dielectric properties were carried out by means of the “Hewlett-Packard – 4192 A” set up, containing thermometer “Keithley-740” and 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 to the sputter target composition, sputtered by 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) had a quasiamorphous structure. Initial structure formation in $PbZr_{0.54}Ti_{0.46}O_3$ takes place at annealing with $T_{anneal} > 450$ °C and $pO_2 = 2\times10^5$ Pa. Most qualitative are films annealed in temperature region $500 < T_{anneal} \leq 550$ ºC.

For the formation of $PbZr_{0.54}Ti_{0.46}O_3$ compounds in ITs of SiO_2 layer the same thermal treatment regimes as those for $PbZr_{0.54}Ti_{0.46}O_3$ films have been used. In the process of study of XPS spectra in concern of 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. 1). The largest maximum of the XPS spectra relates to the structure annealing during 8 min. Corresponding maxima on the XPS spectra were observed for Zr at binding energy of ~182 eV and for Pb at binding energy of ~138 eV.

During measurements of temperature dependencies of dielectric permittivity of Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure in the temperature range of 0-500 °C at various frequencies ($5\times10^3 – 8\times10^5$ Hz) it was determined that a weak increase of ε is observed in the temperature range of 100-250 °C and at low-frequencies (5×10^3 Hz and 4×10^4 Hz). At T > 250 °C a sharp jump of ε up to the $\varepsilon_{max}=4.2\times10^3$ (at T=375 °C and $\nu=5\times10^3$ Hz), $\varepsilon_{max}=4.6\times10^3$ (at T = 380 °C and $\nu= 4\times10^4$ Hz) and $\varepsilon_{max}=5.0\times10^3$ (T=372 °C, $\nu=8\times10^5$ Hz) take place (Fig. 2).

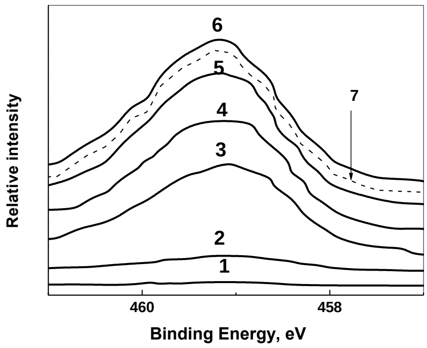

Figure 1. The change of XPS spectra maxima in $PbZr_{0.54}Ti_{0.46}O_3$ compound of the $Si/SiO_2(PbZr_{0.54}Ti_{0.46}O_3)$ structure annealed at 550 ºC and $pO_2=2\times10^5$ Pa for: 3 min (1), 4 min (2), 5 min (3), 6 min (4), 7 min (5), 8 min (6), 9 min (7).

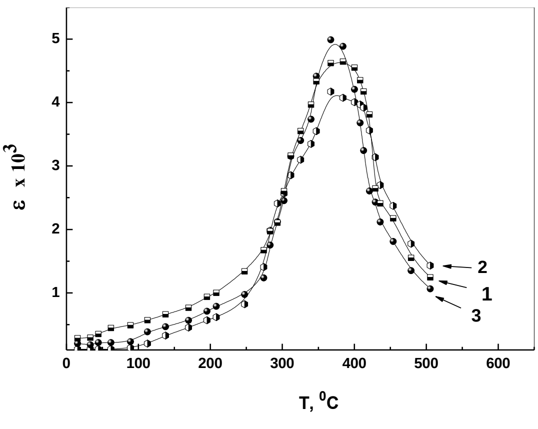

Figure 2. Temperature dependencies of the dielectric permittivity of Si/SiO_2 ($PbZr_{0.54}Ti_{0.46}O_3$) structure at: 5×10^3 Hz (1), 4×10^4 Hz (2) and 8×10^5 Hz (3).

The decrease of ε with ν increase in the temperature range of the existence of the local maximum (ε_{max}) indicates an 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.

If a 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. At the lacking of an interaction between dipoles the disordering effect is not a cooperative one and it does not lead to an occurrence of the FPT. The interdipole interaction lowers the energy of dipole formation with their concentration growth.

4. Conclusion

As a result of the carried out investigations, the following is established:
- chemical deposition of Pb, Zr and Ti metals in swift heavy ion tracks in SiO_2 thin layers on silicon substrates with a subsequent annealing at 550 ºC and $pO_2 = 2\times10^5$ Pa makes it possible to obtain $PbZr_{0.54}Ti_{0.46}O_3$ compounds;
- during investigations of dielectric properties of 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 indicating that ε=f(ν, T) reaches its maximum in the temperature range of 360–406 ºC ;
- maximal dielectric permittivity at 360–406 ºC for various measurement frequencies is attributed to the ferroelectric phase transition in $PbZr_{0.54}Ti_{0.46}O_3$ compound, which leads to a disorder in the cation sublattice and disappearance of ferroelectric properties of the structure.

References

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