DIELECTRIC PROPERTIES OF $Ba_{0.8}Sr_{0.2}TiO_3$ IN HEATING AND COOLING PROCESSES

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The temperature dependences during heating and cooling processes of the dielectric properties of the metal- dielectric-semiconductor (MDS) structure based on ferroelectric $Ba_{0,8}Sr_{0,2}TiO_3$ films deposited on silicon substrates (*n*-type 3KEF and *p*-type 4KDB) by high-frequency sputtering in the crystallographic direction (100) are studied in the temperature range 300-440 K. Several relaxation-type phase transitions associated with the ferroelectric property of these crystals are discovered.

Keywords: ferroelectric film, metal-dielectric- semiconductor, heating, cooling, dielectric constant, dielectric loss tangent.
PACS: 77.80.-e

INTRODUCTION

Currently, one of the promising areas of modern materials science is the creation of new multifunctional materials. Last years, the sharp increase in investigations directed on development of ferroelectric nonvolatile memory with random access [1, 10, 13] has been observed. Such materials have been successfully used in super high frequency (SHF) electronics, including high-voltage pulse technology. In this regard, the use of ferroelectrics as a high-energy-containing materials in the field of electronic devices is relevant. Therefore, the use of ferroelectric films has been recently begun to expand rapidly. It should be noted that in modern microelectronics, SHF electronics is of great practical interest. In this regard, the field of special applications to the category of mass consumption, such as cellular telephony, satellite television, acoustoelectronic devices, etc., is the global information network [2, 8, 11, 12]. Information on the electrophysical properties (C, R, ρ_v, σ, ε, $tg\delta$) of multilayer heterostructures, including high values of the high dielectric constant of the films containing ferroelectrics, including $Ba_{0.8}Sr_{0.2}TiO_3$, plays a particularly important role. Measurements of dielectric properties show that the obtained structures are promising for the construction of spin-wave devices for processing of SHF signals [1, 5, 6, 7, 11]. In this regard, at the investigation of new multifunctional ferroelectric materials it is necessary to have information on the temperature and frequency dispersion of the real (ε') and imaginary (ε'') parts of the complex dielectric constant (ε), the dielectric loss tangent ($tg\delta$), the electrical conductivity at constant (σ_{dc}) and alternating (σ_{ac}) electric fields, and the basic laws of variation of these parameters and the dielectric relaxation spectrum.

It should be noted that in work [2] it was shown that a planar condenser in medium-power SHF devices operates in severe temperature conditions, as well as on the basis of calculations critical operating modes of the capacitor were estimated at frequencies of 3-15 GHz at different powers. The purpose of this work is to study the dielectric properties of $Ba_{0.8}Sr_{0.2}TiO_3$ in heating and cooling processes at a temperature of (293-493K) in alternating electric field.

EXPERIMENTAL TECHNIQUE

As objects of study, we synthesized MDS structures, which are a *p*- and *n*-type silicon substrate, a $Ba_{0.8}Sr_{0.2}TiO_3$ ferroelectric film, and an upper nickel electrode. The thickness of the silicon substrates was 200 ± 2 μm, and the crystallographic orientation was (100). The ferroelectric film was deposited on a silicon substrate by high-frequency sputtering of a polycrystalline target in an oxygen atmosphere using a Plasma-50SE installation. The design of the installation and the technique of film deposition are given in [2].

The upper nickel electrode was deposited onto the ferroelectric film by the electron beam method through a shadow mask. The contacts area was 2.7×10–4 cm2, and the thickness was 0.1 μm. Two MDS structures on p-type silicon (3KDB, 4KDB) type silicon) and two MDS structures on n-type silicon (3KEF, 4 KEF) were fabricated. Under similar technological conditions, a ferroelectric film was applied to the 4 KEF and 4 KDB substrates during 15 minutes, and to the 3 KEF and 3KDB substrates during 20 minutes. The thickness of the ferroelectric film plus the SiO2 insulating layer was 450+10 nm and 480+10 nm, respectively. The electrophysical properties of MDS structures were measured using an E7-20 LCR meter. The frequency range is 25-10^6 Hz [9]. The values of the real and imaginary parts of the dielectric constant were calculated based on the thickness of the ferroelectric film d and the contact area S according to the formulae for a flat capacitor $\varepsilon'=c\cdot d/\varepsilon_0 S$ and $\varepsilon''= tg\delta\varepsilon'$ at the voltage U=1V.

EXPERIMENTAL RESULTS AND THEIR DISCUSSION

It is known that mechanical stresses can significantly affect on the temperature dependences of the dielectric constant ε (T) and the dielectric loss tangent $tg\delta=f(T)$.

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It is shown that the obtained structures are promising for the construction of spin-wave devices for processing of SHF signals. It should be noted that each ferroelectric material has a number of peculiar electrophysical properties. It is known [1, 4, 7, 14] that ferroelectrics are called polar dielectrics, which in a certain temperature range possess spontaneous polarization. Obtaining of ferroelectric materials is one of the important tasks to predict the effects of external factors (external electric fields, temperatures, pressure, humidity, ionizing radiation, etc.). Fig. 1-4 shows the dependences $\varepsilon'=f$(T) and $tg\delta=f$(T) at heating and cooling of 3KEF and 4 KDB samples.

In [3, 4] it is shown that various scattering processes make contributions to the value of tgδ; these contributions are important in the research of low-loss microwave dielectrics. This makes it possible to use these ferroelectrics in microwave electronics, including high-voltage pulse technology. Fig. 1 shows the $\varepsilon'=f$(T) dependence of 3KEF sample with MDS structure during heating and cooling.

As seen in the process of heating (fig. 1. 3KEF), with increase the of the temperature there is observed an increase in the value of ε', spontaneous polarization is observed in the polar phase at temperatures below the Curie temperature. At T=393K, the Curie temperature is maximized and the phase transition is manifests itself. Let's also note that, regular and irregular transitions, typical for the ferroelectricity depend on the structure elements (molecules, ions and radicals) of the crystal and these structure elements may be in two or more equilibrium states. It's on its turn characterized by a dipole moment. In the temperature above the Curie temperature in the nonpolar phase, ε', along with the dependence from the temperature, also depends on the field voltage. In the high-temperature area (after the temperature of the Curie), with increasing of energy, the dipole-dipole interaction increases as a result of the heat flux and every instant, the dipoles are randomly directed, as a result, the total polarization equals to zero (total dipole moment P=0) and ferroelectricity in the non-polar phase behaves like a dielectric. Now, if we look at the reverse process, we will see that with decreasing of the temperature due to dipole-dipole interactions in the phase transition, the formation of elements with a regular polar structure takes place in itself, therefore, spontaneous polarization and P>0 occur in the ferroelectric samples under study. Note that a phase transition at T=393K also occurred during the cooling process.

Temperature dependence of tangent of dielectric loss angle ($tg\delta=f$(T)) on the heating and cooling processes is given in fig. 2. Here if we see curve 2, we can observe creation of several maxima (T=363K, T=393K) and this is can be connected with several Curie temperatures, which is characteristic with ferroelectrics. The dependences $\varepsilon'=f$(T) and $tg\delta=f$(T) of the 4KDB sample in heating and cooling processes are given in fig. 3 and fig. 4. Here the same tendency manifests itself in the same way as the previous explanation. Here, also in the curve in fig. 3, depending on the temperature the formation of several maxima is observed, which also characterizes the phase transitions arising from ferroelectric properties and being of the process - relational type. This is characteristic of the dependence of the electrophysical properties of the MSD crystalline system on both temperature and frequency.

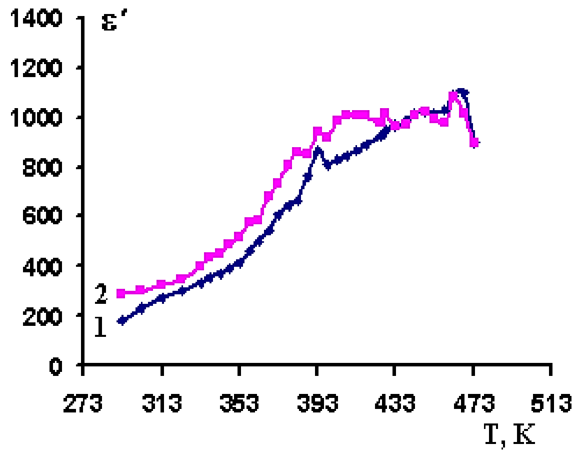

Fig. 1. $\varepsilon'=f$(T) dependence for 3KEF in heating and cooling processes. 1 is heating, 2 is cooling.

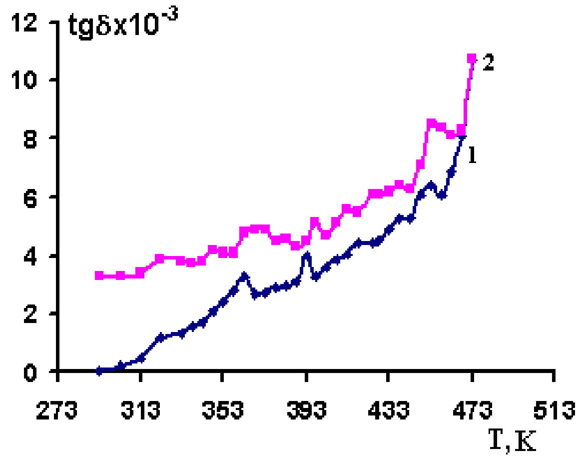

Fig. 2. $tg\delta=f$(T) dependence of 3KEF in heating and cooling processes. 1 is heating, 2 is cooling.

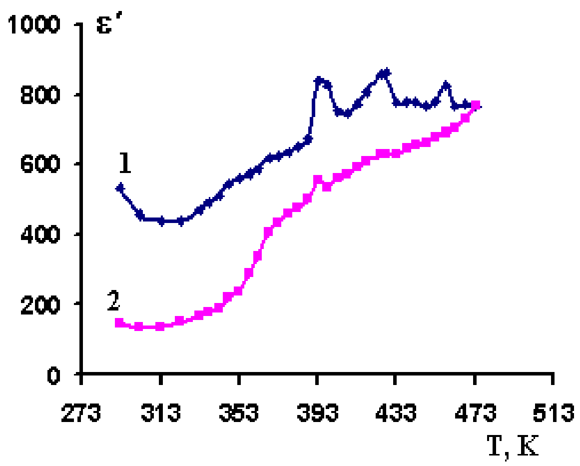

Fig. 3. $\varepsilon'=f$(T) dependence of 4 KDB in the heating and cooling processes. 1 is heating, 2 is cooling.

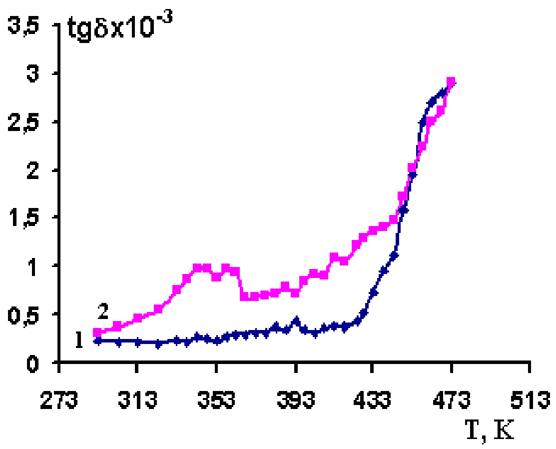

Fig. 4. tgδ=f(T) dependence of 4KDB in heating and cooling processes. 1 is heating, 2 is cooling down.

Temperature dependence of the activation energy of relaxation processes during heating and cooling processes of 3KEF and 4KDB samples at 1 kHz is calculated with Arrenius formula

$$E_{ak}=kT\ln(\tau/\tau_0) \quad (1)$$

here, τ_0 is characteristic relational time in the same arrangement with the dance phase of the atom ($\tau_0 \sim 10^{-13}$ sec.), τ is the duration of the external area and $\tau=10^{-3}$ sec [15]. The activation energy of the relaxation process during the heating process increases from 1,02eV to 1,65eV in the temperature range 293÷473K. In the reverse cooling process the activation energy decreases.

As it is seen in fig. 1-4 during heating and back cooling processes the shifted maxima of the relaxation process according to the corresponding temperature are observed

An increase in the activation energy upon heating and a decrease upon cooling is a common regularity for the measured dependences, this is also associated with a change in carriers energy due to temperature changes and so mean corresponds to a change in the Fermi status within the band gap of the material. Changes in the dielectric properties of this medium can lead to the release of trapped loads. On the other hand, a breakthrough may occur for new energy states in a MDS medium.

It should also be noted that as the temperature rises, the mobility of the structural elements of the samples also increases. The higher the molecular velocity, the greater the movement and turning angle of the domains in the electric field. Consequently, the dependence of the electrical conductivity and the tangent of the loss angle associated with it, the dielectric relaxation time and the activation energy of polarization processes at certain areas of the temperatures can be stepwise, what is seen in fig. 1-4. Here, on the curve of the cooling down process, the stepwise character is more noticeable (fig. 1-4, curve 2), this behavior can be explained by the phase transition in the material, i.e., the transition from the ferroelectric phase to the paraelectric phase.

CONCLUSION

The results of the carried investigations show that in temperature dependence of dielectric properties of crystal with MDS structure (3KEF and 4KDB) the several phase transitions are observed. It is connected with the fact that these crystals have the ferroelectric properties.

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Received: 13.03.2020

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