

Nanoscale barium-strontium titanate films for microelectronics devices

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The temperature dependence of permittivity and leakage currents of nanoscale thin films of barium-strontium titanate are investigated. The studies were carried out with a bias voltage applied to the gap between the pins of an interdigital capacitor based on the specified film. High temperature stability of nanoscale thin films of barium-strontium titanate with bias voltage applied is established. It is shown that leakage currents are nonlinear and asymmetrical relative to the applied bias voltage, and their value allows using the studied films in the implementation of microelectronic devices.

Keywords: nanoscale, ferroelectric film, planar capacitor, barium-strontium titanate.

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The progress in microelectronics stimulates both the search for new active media and further improvement of time-tested techniques used to fabricate components for electronic devices and systems based on them.

Integrated device design, which allows one to fabricate the entire structure in a single technological cycle, is an essential condition for inexpensive mass production. It is the reduction of cost of manufactured devices that stimulates the use of integrated technology. The key trends in evolution of elements forming the hardware base of integrated circuits are the reduction of overall dimensions, surface mounting without any external components, expansion of the operating frequency range, and enhancement of thermal stability and radiation resistance. The listed trends have led to the development of new elements: thin-film heteroepitaxial structures. This was made possible, among other things, by the use of new active media. Although the attention of researchers and engineers has recently been focused on such new active media as, e.g., graphene and liquid crystals [1,2], studies into the feasibility of application of ferroelectric materials in microelectronics remain relevant [3]. In particular, nanoscale ferroelectric films continue to attract research attention, since they offer certain hitherto unmatched characteristics [4]. Specifically, liquid crystals used as an active medium in microwave devices cannot provide a speed performance comparable to that of thin ferroelectric films, since it takes a relatively long time for extended molecules of a liquid crystal dielectric to reorient under the influence of an electric field [5]. Another limiting factor is the operating temperature of devices with liquid crystals (from -25 to $+55$ °C) [5]. Owing to the complexity of the technological cycle (including, e.g., thermal annealing for removal of impurities from the surface) [6], graphene turns out to be inferior to nanoscale ferroelectric (FE) films in the case of mass production of integrated circuits designed for use in microwave devices. Thin ferroelectric films for real-world applications need

to provide high temperature stability within a wide temperature range and (ideally) the weakest possible leakage currents under the application of a bias voltage. This necessitates a thorough study of thermal stability and leakage current characteristics in thin ferroelectric films. These issues have been discussed in detail in [7,8] for thin ferroelectric films of lead zirconate titanate formed by the sol-gel method. The issue of thermal stabilization of multilayer capacitive metal/ferroelectric/metal structures based on barium strontium titanate films of various compositions with a thickness of several hundred nanometers has been investigated thoroughly in [9].

The aim of the present study is to examine the dependence of permittivity on temperature (thermal stability) and to estimate the magnitude of leakage currents of nanoscale (thinner than 50 nm) ferroelectric $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ films with a bias voltage applied to them. The goal is to use the obtained data to assess the feasibility of application of such films in the production of basic microwave elements. Nanoscale ferroelectric films of the $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ composition and interdigital capacitors, which are one of the key basic microwave elements, fabricated with the use of these films were studied. The examined films were thinner than 50 nm and were synthesized using a „PLASMA-50 SE“ setup by sputtering a stoichiometric $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ target with an asymmetric capacitive radio-frequency (RF) discharge. The deposition of films onto a magnesium oxide (MgO) substrate 500 μm in thickness was carried out in an atmosphere of pure oxygen at a pressure of 0.45 Torr and a temperature of 700 °C. The need to obtain single-crystal $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ films of high structural perfection during epitaxial film growth motivated our choice of MgO single crystals as substrates. The inevitable presence of crystallites in nanoscale $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ films formed by RF deposition with conventional microelectronic substrates (silicon, sapphire, polycor, sitalls) leads to a significant increase in dielectric losses. We used commercially available

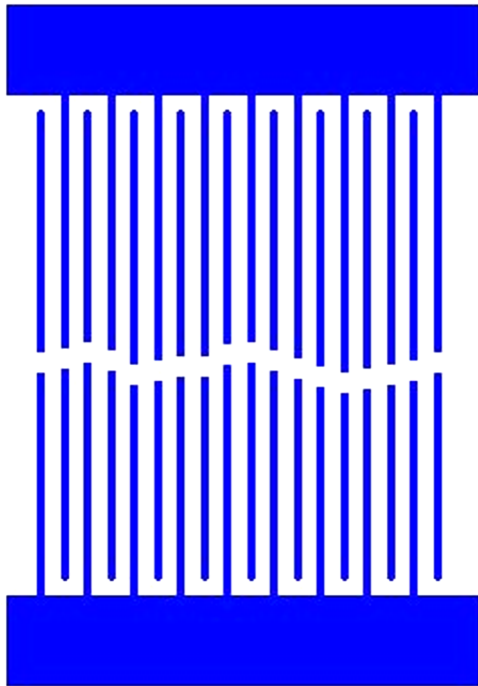


Figure 1. Interdigital capacitor topology.

MgO single-crystal substrates with a permittivity of 9.6. The element topology was formed by lift-off photolithography. To that end, a photoresistive mask was formed on the film surface, and an aluminum film with a thickness of $0.3\mu\text{m}$ was deposited onto it by magnetron sputtering. At the second stage, the basic topology of the device under study with an aluminum film thickness of $2.0\mu\text{m}$ was formed.

Planar capacitors play a key role in the design of most basic elements used in systems and complexes operating in the microwave range. The preferred design of a planar capacitance is an interdigital capacitor (see its topology in Fig. 1).

Its key advantages are the ease of manufacture and a high Q factor.

We have built capacitors of this type based on films of the $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ composition with a nanoscale thickness. The films were deposited onto magnesium oxide substrates using the method described above. The film thickness was 21 nm. The interdigital capacitors had the following dimensions: gap width, $1.38\mu\text{m}$; pin width, $2\mu\text{m}$; pin length, $310\mu\text{m}$. The structural perfection of films, lattice cell parameters in the direction normal to the substrate plane and along the substrate plane, and epitaxial relations between the film and the substrate corresponding to different growth mechanisms were determined by X-ray diffractometry using a DRON-4-07 setup (recording of symmetric and asymmetric Bragg reflections). The survey X-ray diffraction pattern obtained by scanning within the range of angles 2θ from 20 to 140 deg revealed only 001-type reflections of the film (Fig. 2) and the substrate, which indicates that the [001] axis of the film is oriented parallel to

the [001] axis of the substrate. The bulk $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ (BST) sample has tetragonal symmetry of the lattice cell with lattice parameters $a = 0.398\text{ nm}$, $c = 0.399\text{ nm}$. The lattice cell parameter in the direction perpendicular to the substrate in the $\text{MgO}/\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ heterostructure is $c = 0.3988\text{ nm}$. The lattice cell parameter in the substrate plane for $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ is $a_{0.8} = 0.4001\text{ nm}$; i.e., it is larger than the parameter along the normal to the substrate. This suggests that the single-crystal film in the $\text{MgO}/\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ heterostructure is subject to two-dimensional tensile stresses. The epitaxial nature of growth of all studied films was verified by the results of φ -scanning of asymmetric 113 and 103 reflections of the film and 113 reflections of the substrate. A parallel arrangement of axes of the film and the substrate in the interface plane was typical of the $\text{MgO}/\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ heterostructure; i.e., $[100]\text{BST} \parallel [100]\text{MgO}$, $[010]\text{BST} \parallel [010]\text{MgO}$, and $[001]\text{BST} \parallel [001]\text{MgO}$. The vertical and azimuthal misorientations of the films were less than 0.4° .

The temperature characteristics of films and the dependence of the leakage current density on the bias voltage applied to the interdigital capacitor were investigated.

Capacitance–voltage characteristics were measured using an instrumentation complex based on an Agilent 4980A precision LCR meter. To carry out temperature measurements, the sample holder was introduced into a thermostat allowing for both heating and nitrogen vapor supply from a Dewar vessel.

The dispersion of dielectric characteristics of thin ferroelectric films may be indicative of their low quality. It was proven in [10] that $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ films of a nanoscale thickness have almost zero dispersion of permittivity ϵ' and an insignificant (at least in the microwave range) dispersion of dielectric losses ϵ'' at frequencies up to the terahertz range. This allows us to extrapolate the results of low-frequency capacitive measurements for the formed structures to the microwave range (at least up to the W band). According to the data reported in [10],

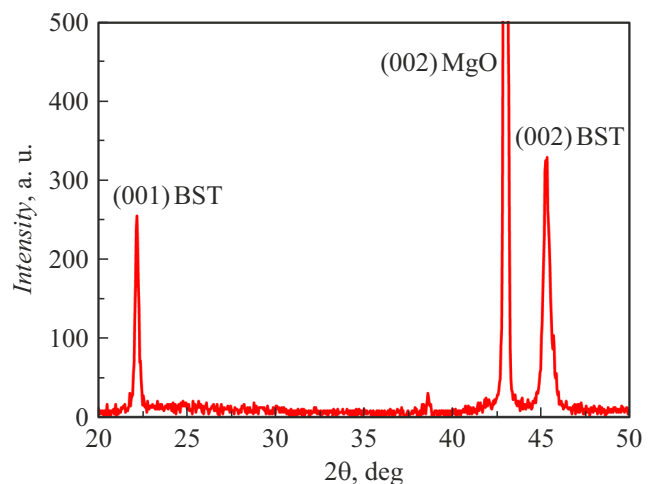


Figure 2. Fragment of the X-ray diffraction pattern in the region of reflections 001 and 002 of the MgO/BST heterostructure.

the magnitude of dielectric losses ($\tan\delta$) for films with a nanoscale thickness does not exceed 0.01 in the microwave range.

It follows from Fig. 3 that the measured temperature dependence of the dielectric characteristics of the studied nanoscale $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ films in the ferroelectric phase revealed their high temperature stability under the application of a bias voltage.

An exhaustive explanation of the phenomenon of thermal stability of a varactor based on two thin layers of barium strontium titanate of different compositions was provided in [9]. The varactor studied in [9] was specific in that it used 200–600-nm-thick $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ films with the composition (x) varying along their thickness. Such films may be regarded as a multilayer composition with different phase transition temperatures of each layer. A definitive interpretation of the experimental results we obtained, which are indicative of stable temperature behavior of permittivity of the studied heterostructure based on nanoscale films under the application of a bias voltage, has not been formulated yet and evidently requires a more thorough study of the physical aspects of this phenomenon. One peculiar feature of the examined heterostructure is „blurring“ of the phase transition for $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ films of nanoscale thickness, which is attributable to the size effects and the specifics of internal mechanical stresses. In the vicinity of the phase transition point, the system is most unstable with respect to external influences. The stability increases with „blurring“ or with distance from the phase transition point. Applying a bias voltage to the FE film causes a change in „soft mode“ frequency, which leads to a reduction in permittivity. The film permittivity reaches its maximum near the point of phase transition (Curie temperature) from the ferroelectric state to the paraelectric one. However, it is precisely in the vicinity of this point that the maximum instability under external influences is observed. Thus, the reduction in permittivity under the application of a bias voltage transfers the system to a state of greater stability, which covers, among other

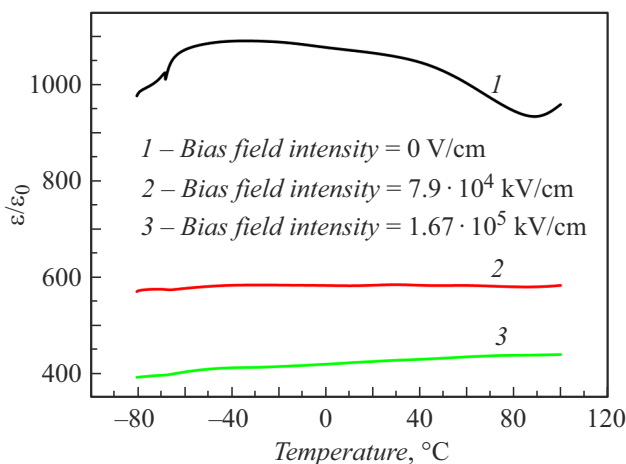


Figure 3. Temperature dependence of the film permittivity.

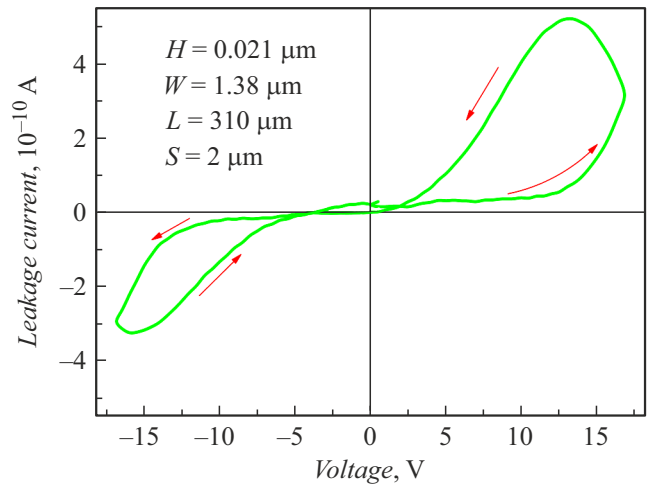


Figure 4. Dependence of leakage currents for the interdigital capacitor.

things, the resistance to temperature changes. And this is the very definition of thermal stability: the capacity to maintain constant physical properties under temperature variations.

The leakage currents of interdigital capacitors were measured using a Keithley 4200SCS parameter analyzer with a PM-5 MicroTec probe station at a maximum control voltage of 20 V. These currents did not exceed $5 \cdot 10^{-10}$ A, and their maximum density was below $4 \cdot 10^{-5}$ A/cm².

It follows from the data presented in Fig. 4 that the leakage currents within the operating range of bias voltages $V = 1\text{--}20$ V are nonlinear and asymmetrical with respect to the applied bias. The experimental dependence has a „hysteretic“ character.

The obtained results demonstrated that the thermal stability and leakage currents of nanoscale barium strontium titanate films are suitable for their application in basic microwave elements that may be used successfully in the engineering of microwave devices and systems of an integrated design.

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Conflict of interest

The authors declare that they have no conflict of interest.

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