Piezoelectric properties of $SrBi_2(Ta_xNb_{1-x})_2O_9$ thin films synthesized by the sol-gel method

A. V. SEMCHENKO[1], V. V. SIDSKI[1], O. I. TYULENKOVA[1], A. YU. SHEPELINA[2], D. A. KISELEV[2], V. A. PILIPENKO[3]

[1]Francisk Skorina Gomel State University, Sovetskaya 104, Gomel, 246028, Belarus
[2]National University of Science and Technology “MISiS”, Leninsky pr.4, Moscow,119049, Russia
[3]JSC “INTEGRAL”, Kazintsa I.P 121A, Minsk, 20108, Belarus

Abstract
The present work aims to design and study novel functional thin films with ferro- and properties. $SrBiTa_2O_9$ and $SrBi_2(Ta_xNb_{1-x})_2O_9$thin films were synthesized by the sol-gel method on $Pt/Ti/SiO_2/Si$ substrates. The influence of the synthesis conditions and the presence of niobium on the features of the piezoelectric properties were determined.The PFM method was applied to visualize not only the morphology of grains, but also their local piezoelectric activity.

Introduction

At the moment, dielectrics and, in particular, ferroelectrics in the form of thin films, which have the greatest prospects for practical applications for miniaturizing devices, due to reduced control fields, good integration of films with semiconductor materials; traditionally used in electronics, as well as for creating reprogrammed memory devices based on ferroelectrics [1]. Ferroelectrics are characterized by the presence of dielectric hysteresis loop, large dielectric constant, high piezomodule, as well as some electro-optical properties. The main practically used property of such materials is their controlled dielectric response to external influence, which, as a rule, is based on polarization switching processes. The widespread practical use of ferroelectric materials is constrained by the incomplete research on polarization switching dynamics in these structures. Local piezoelectric properties of $SrBiTa_2O_9$ (SBT) and $SrBi_2(Ta_xNb_{1-x})_2O_9$ (SBTN) thin films are investigated in this work.

Experimental

The main problem in the formation of a homogeneous multicomponent solution is the unequal conditions of hydrolysis and the rate of condensation for each type of metal alkoxides. This can lead to phase separation during hydrolysis or heat treatment and, consequently, to crystallization temperature increase or even to the formation of impurity phases. Thus, choosing such alkoxides and a solvent with sufficient solubility is necessary for the stoichiometric ratio in their joint presence and do not interact at room temperature. Many problems of the above-described variant of preparing a mixture of a working solution can be overcome by replacing the alkoxides of some metals with their salts. These are mainly salts of carboxylic acids, but nitrates, sulfates and chlorides can also be used. The salts of acetic acid and ethyl caproic acid are versatile and can be used for most metals. They are compatible with almost all metal alkoxides. It was found that the use of separate hydrolysis improves the ferroelectric properties of the obtained films. This is due to the fact that when preparing the sol when using the same solvent, separate hydrolysis occurs completely for each of the metal compounds in comparison with joint hydrolysis, where, due to different reaction rates, some metals are not completely hydrolyzed.
Synthesis of $SrBiTa_2O_9$ films used nitrate salts of Sr, Bi, Ta, HNO_3, ethylene glycol, ethylenediamine and citric acid. The $SrBi_2(Ta_xNb_{1-x})_2O_9$ films were synthesized using the same procedure. $NbCl_5$ was used as a niobium source. The thickness of the films can be controlled by the number of layers spin coated. Then the samples $SrBiTa_2O_9$ and $SrBi_2(Ta_xNb_{1-x})_2O_9$ sol-gel materials were annealed at different temperatures for 20 minutes.

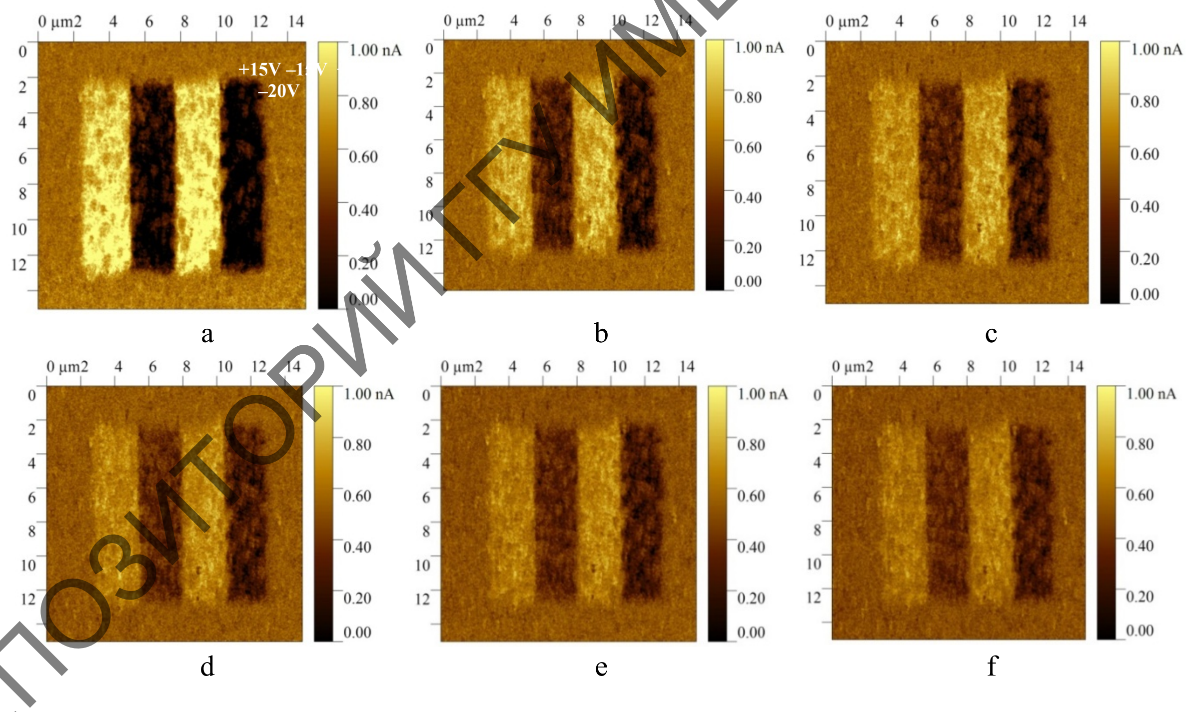

Fig. 1. Polarization of SBTN film with annealing temperature 700 ° C after polarization with voltage ± 15 V and ± 20 V (in contact mode): a – immediately after polarization; b – 10 minutes after polarization; c – 20 minutes after polarization; d – 30 minutes after polarization; e – 40 minutes after polarization; f – 50 minutes after polarization

Results and discussion

The vertical or out-of-plane component of the piezoresponse was measured, which is proportional to the effective piezoelectric effect d_{33} [1]. The images showed SBTN thin films annealed at different temperatures. The convenient way to describe a piezoresponce is to construct it in the form of histograms as the function of the image of pixels in the entire image [2]. In this way, complete polarization distribution in the resulting films can be obtained. Due to the small grain size, the contrast in the grain is constant, and no ferroelectric domains are observed.

Visualization of the induced domain state is possible after polarization of the film by applying constant voltage to the "cantilever - film - substrate" system. For this, the surface of the film, measuring (2.5x10) μm^2, was first polarized with a constant voltage of ± 15 V and ± 20 V, and then a larger area of the film was scanned in the piezoresponse mode (15x15) μm^2. The results are shown in Figure 1. It was established experimentally that the best "memory" for polarization is a sample with niobium content of 40%.

Conclusion

Polycrystalline $SrBi_2(Ta_xNb_{2-x})O_9$ (SBTN) films (700 °C annealing temperature) with different Nb content from 10% to 50%, with a step of 10%, as well as 5 samples of SBTN films (20% content Nb) were synthesized. It was experimentally established that the best "memory" for polarization is a sample with niobium content of 40%.

References

[1] P. K. Panda and B. Sahoo. Ferroelectrics, (2015) 474, 128 – 143.

[2] P. Sharma, Q. Zhang, D. Sando, C. H. Lei, Y. Liu, J. Li, V. Nagarajan, J. Seidel, Nonvolatile ferroelectric domain wall memory. Sci. Adv. 3, e1700512 (2017).