Electrical properties of $Na_{0.5}Bi_{0.5}TiO_3$ thin films deposited on sitall substrates.

Kruzina T,V., Popov S.A., Potapovich Yu.N., Ryabtsev S.I., Rutskiy A.S.

Experimental Physics dept., Oles Honchar Dnipro National University ,
prosp. Gagarina 72, Dnipro, Ukraine, 49010 e-mail: spopov@meta.ua

Introduction

Ferroelectric $Na_{0.5}Bi_{0.5}TiO_3$ (NBT) is a promising material to use in electronic engineering. At present, the best samples of obtained thin NBT films show the dielectric constant of 600–800 and the remnant polarization of 19 $\mu C/cm^2$. Despite some technological achievements in preparation of NBT films, the relationship between their electrical properties and such factors as technological parameters, the substrate material, as well as the material of the lower electrode and the state of its surface has not been studied sufficiently. Such important disadvantage of NBT films as presence of significant leakage currents (~$10^{-7} A/cm^2$) [1,2] can also be influenced by abovementioned factors. Although most often NBT films are formed on silicon substrates, in this work we investigate electrical properties of NBT films deposited by the RF magnetron sputtering on sitall substrates. Such substrates have high values of bulk and surface resistivities, low values of dielectric losses, have high thermal conductivity, and what is very significant they have small values of linear expansion coefficients.

Materials and Methods

The NBT films were obtained by ion-plasma magnetron high-frequency (RF=13.56 MHz) methods of sputtering of ceramic NBT target in an argon atmosphere (chamber pressure 1 Pa). The target was prepared according to the technology described in [3]. Sitall substrates were chemically and ionically cleaned in advance. A Pt layer (of 85 nm thickness) was deposited on the clear substrate by DC magnetron sputtering. The crystallographic structure of the Pt layer would promote formation of a correct NBT films structure and the layer was also used as the lower electrode. The NBT film was deposited on the Pt covered substrate heated to 150°C, the temperature that was optimal given the film composition contained the volatile components. Subsequent synthesis of the films was carried out in an unsealed container with coarse crystalline NBT powder at temperature around 550°C-700°C for 45 minutes in air. The excessive vapor pressure of the volatile Na and Bi components compensated for the violation of the film stoichiometry during the synthesis. The phase composition of obtained films was controlled by X-ray phase analysis. The leakage current-voltage (I-U) characteristics were measured by voltmeter-electrometer. The remnant polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) were measured at room temperature by modified Sawyer-Tower circuit.

Experimental results

Immediately after the deposition process the NBT films were amorphous. Heat treatment of amorphous NBT at 550°C leads to formation of an intermediate low-temperature pyrochlore phase [2]. The XRD patterns of the NBT films annealed at 700°C showed presence of the main attribute reflections of the perovskite phase (2Θ=23°15'(100), 2Θ=33°20'(110), 2Θ=40°25'(111)) and indicated formation of the ferroelectric NBT phase.

Ferroelectric properties of the NBT films are confirmed by hysteresis loop. The Pr, Ps, Ec values are 9 $\mu C/cm^2$, 15 $\mu C/cm^2$, 38 kV/cm, respectively (Fig. 1). The dielectric constant ε measured at 1 kHz was ~ 720.
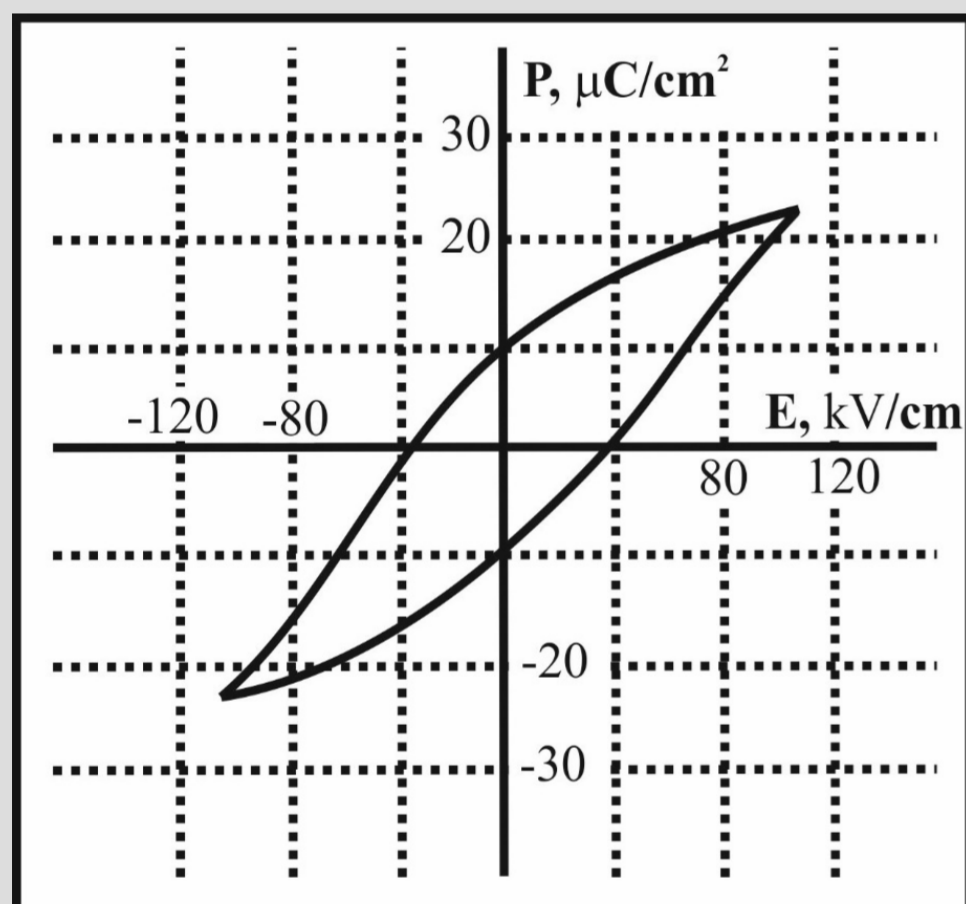

Fig.1. P-E curve of NBT/sitall thin films

Figure 2 shows the dependence of leakage current density on applied electric field (J-E) in NBT films deposited on different substrates: Pt/sitall (NBT/sitall films) and $Pt/TiO_2/SiO_2/Si$ (NBT/Si films).
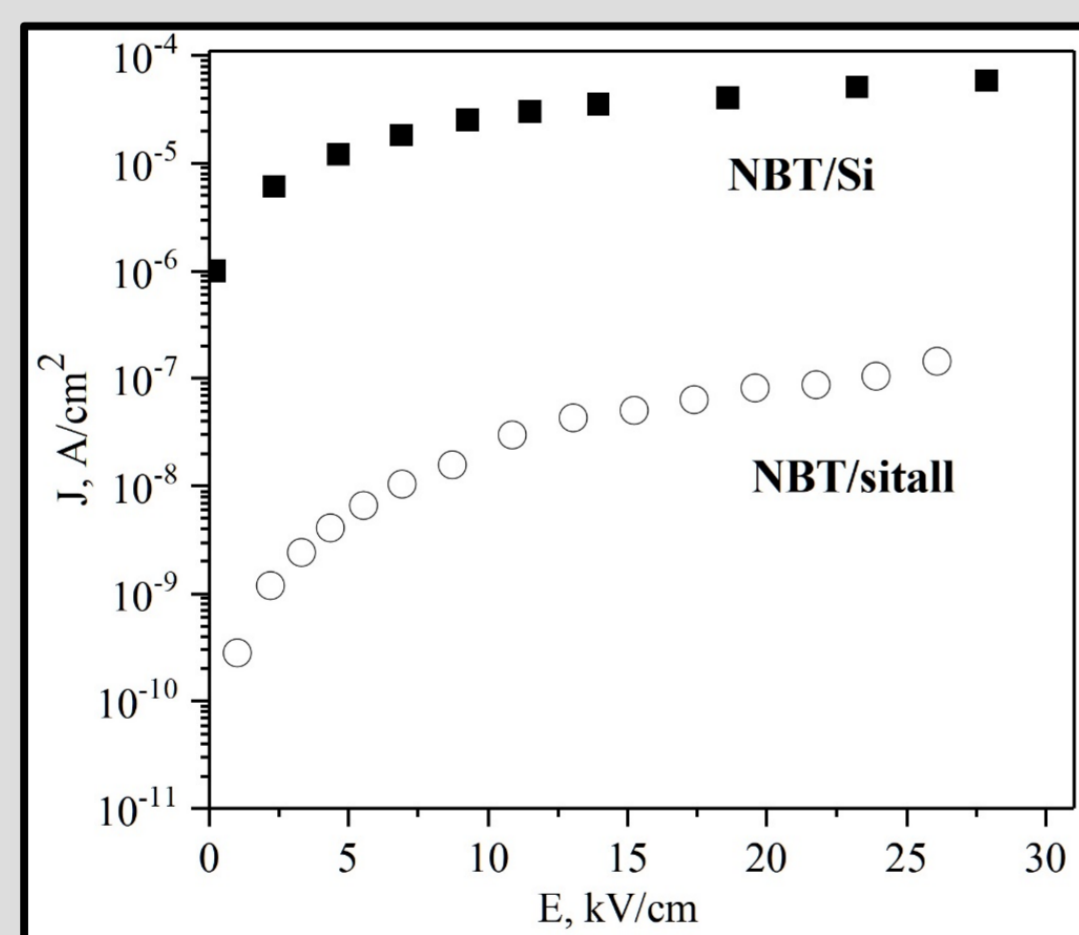
Fig.2. The leakage current density of thin films NBT as function of applied electric field

The significant reduction of the current density in the polycrystalline NBT/sitall films in comparison with the NBT/Si most likely can be associated with changes of the film-substrate boundary morphology.
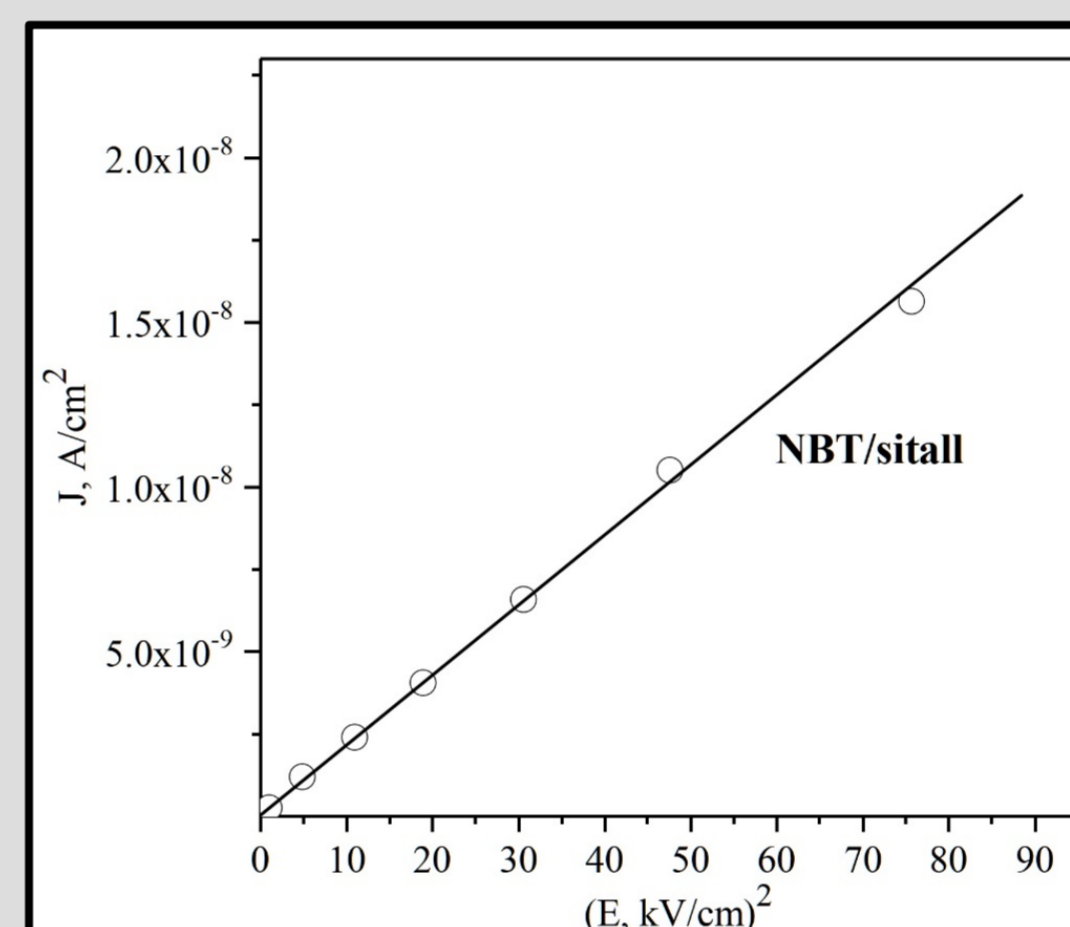

Fig.3. The leakage current density of thin films NBT/sitall as function of E^2

For fields E > 10 kV/cm the most probable mechanism of conductivity is the Schottky emission. Figure 4 shows how the current density depends on the applied field in the Schottky representation. The $ln(J)-E^{1/2}$ dependence seems to be linear, which indicates that the contribution of the Schottky emission charge carriers to the leakage current is predominant.

To determine the conductivity mechanism in the samples of NBT films deposited on the Pt/sitall structure the experimentally measured leakage current data were replotted according to various transportation mechanisms. Fig. 3 shows the $J-E^2$ dependence for the NBT films. The linear form of the curve on the $J-E^2$ graph indicates that the space-charge-limited conductivity dominates in the range of weak fields (E < 10 kV/cm).
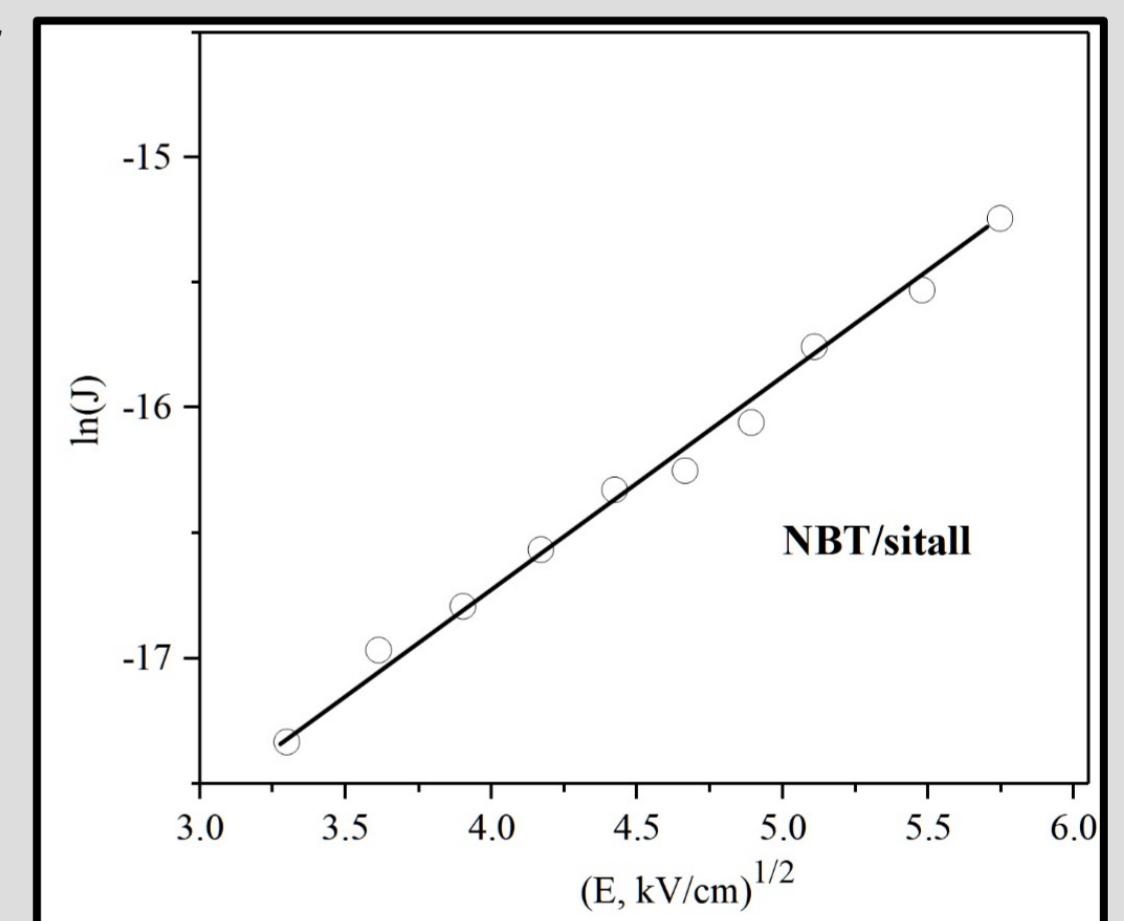

Fig.4. Schottky representation for thin films NBT/sitall

It should be noted that electrical conductivity of polycrystalline films is influenced by the presence of grains and grain boundaries, as well as structural defects including such defects as the oxygen vacancies V_{O_2}-··. In addition grain boundaries in polycrystalline films are preferred sites for formation and accumulation of defects. Oxygen vacancies localized at the grain boundaries can increase leakage currents of NBT films [1, 2].

Conclusions

1. Observed P-E dielectric hysteresis loops confirmed that the polycrystalline NBT films deposited on the Pt/sitall substrates had ferroelectric properties.

2. The space-charge-limited conductivity mechanism dominated in the obtained NBT films in fields E <10 kV/cm. For fields E >10 kV/cm the most probable conductivity mechanism is the Schottky emission.

3. The presence of the oxygen vacancies V_{O_2}-·· allows to suggest that the hopping conduction mechanism may also contribute to leakage currents in NBT films.

4. The significant decrease of the current density in the polycrystalline NBT/sitall films in comparison to the NBT/Si most likely can be associated with morphology changes on the film-substrate boundary which lead to reduced number of structural defects, including V_{O_2}-··, influencing the leakage currents.

References

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