STRUCTURAL AND MORPHOLOGICAL PROPERTIES OF $Sr_{0.5}Ba_{0.5}Nb_2O_6$ THIN FILMS DEPOSITED BY RF-PLD TECHNIQUE ON Pt:Si AND MgO SUBSTRATES

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Epitaxial thin films of strontium barium niobate $Sr_{0.5}Ba_{0.5}Nb_2O_6$ (SBN:50) are deposited on Pt:Si (111) and MgO (001) substrates by radio-frequency assisted pulsed laser deposition technique. We reported the influence of thermal contact conditions of the substrates during deposition on growth of SBN:50 thin layers. For unsecured thermal contact of substrates, the morphology of SBN:50 thin films show large grains and relatively high roughness. The XRD analyses indicate that the highly c-axis oriented SBN:50 thin films are obtained for substrates with high thermal contact.

Keywords: SBN ceramic target, SBN thin films, pulsed laser deposition (PLD), radio-frequency assisted pulsed laser deposition (RF-PLD)

1. Introduction

Strontium barium niobate $Sr_xBa_{1-x}Nb_2O_6$ ($0.25 \leq x \leq 0.75$, SBN) is a ferroelectric solid solution between $SrNb_2O_6$ and $BaNb_2O_6$ with tetragonal tungsten bronze (TTB) type structure [1, 2]. SBN exhibits an exceptionally large electro-optic (EO) coefficient, r_{33} which can range from 400 ÷ 1350 pm/V, much higher than $LiNbO_3$ ($r_{33} \sim 31$ pm/V) the current standard electro-optic material in industry [3-5].

For many applications as pyroelectric detectors, electro-optic modulators or surface acoustic waves devices the material is used in the single crystal form. This makes it expensive and difficult to growth. In this case, the production of SBN epitaxial thin films represents an alternative for SBN single-crystal [3].

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Pulsed laser deposition (PLD) is one of the most attractive techniques for obtaining SBN thin films on different substrates. In other works [6-9] it was reported the using of GaAs, $SrTiO_3$, MgO or LNO coated MgO substrates for the epitaxial growth of SBN thin layers.

Radio-frequency (RF) discharge in oxygen was demonstrated the improving of oxygenation in the thin layers during deposition by PLD technique [10].

In this work, we report on the obtaining of SBN:50 thin films on Pt:Si (111) and MgO (001) substrates by RF-PLD (radio-frequency assisted pulsed laser deposition) technique using a stoichiometric SBN:50 ceramic target. The thermal contact between the substrate and the heater was proving to have significant influence over structural and morphological properties of SBN thin films.

2. Experimental

SBN:50 thin films have been obtained using the RF-PLD technique. The beam from a Surelite II Nd:YAG laser working at 265 nm wavelength with pulse length of 5 ns and repetition rate of 10 Hz was used as irradiation source. For growth of SBN:50 thin layers the commercial Pt:Si (111) and MgO (001) were used as substrates. Polycrystalline SBN:50 ceramic target was prepared by solid-state reaction method. The green body of SBN:50 composition was sintered in air at 1350^{o}C for 10 h in order to obtain a good densification. The experimental procedure of obtaining SBN:50 ceramic target was discussed in a previous paper [11]. For obtaining epitaxial SBN:50 thin films the RF-PLD experiments have been done using the following experimental parameters: the SBN:50/Pt:Si (111) thin film have been obtained using a laser fluence of 2.0 J/cm^2 at 0.6 mbar O_2 partial pressure. The deposition temperature has been set at 730^{o}C, while the substrate was bonding on the heater with silver paste. In the case of SBN:50/MgO (001) thin films, in order to study the influence of the thermal contact between the substrate and the heater, two samples have been obtained at a deposition temperature of 650^{o}C, with the substrate bonded on the heater with silver paste and without silver paste - the substrate was mechanically fixed on the heater. Additionally, a sample at higher deposition temperature of 700^{o}C without a secure thermal contact (no silver paste) has been deposited, while keeping constant the other experimental parameters (laser fluence 2.0 J/cm^2 and O_2 gas deposition pressure at 0.6 mbar). The radio-frequency power during deposition and for the cooling procedure was 100 W. In order to achieve a uniform ablation, the SBN:50 ceramic target was simultaneously rotated and translated. The morphology of the surfaces was studied with an XE 100 Atomic Force Microscope from Park Systems. The surface features and the roughness (RMS) of the films surfaces were measured. The crystalline phase composition was analyzed by X-ray diffraction on an Panalytical X'Pert PRO MRD system in Bragg-Brentano geometry.

3. Results and discussion

3.1. Surface morphology

The influence of the thermal contact conditions of the substrates during deposition on growth of SBN:50 thin films and morphology development was revealed by AFM investigation. All samples show uniform, nanostructured morphologies, without droplets and cracks, with grain sizes varies between 120 and 300 nm (Fig. 1).

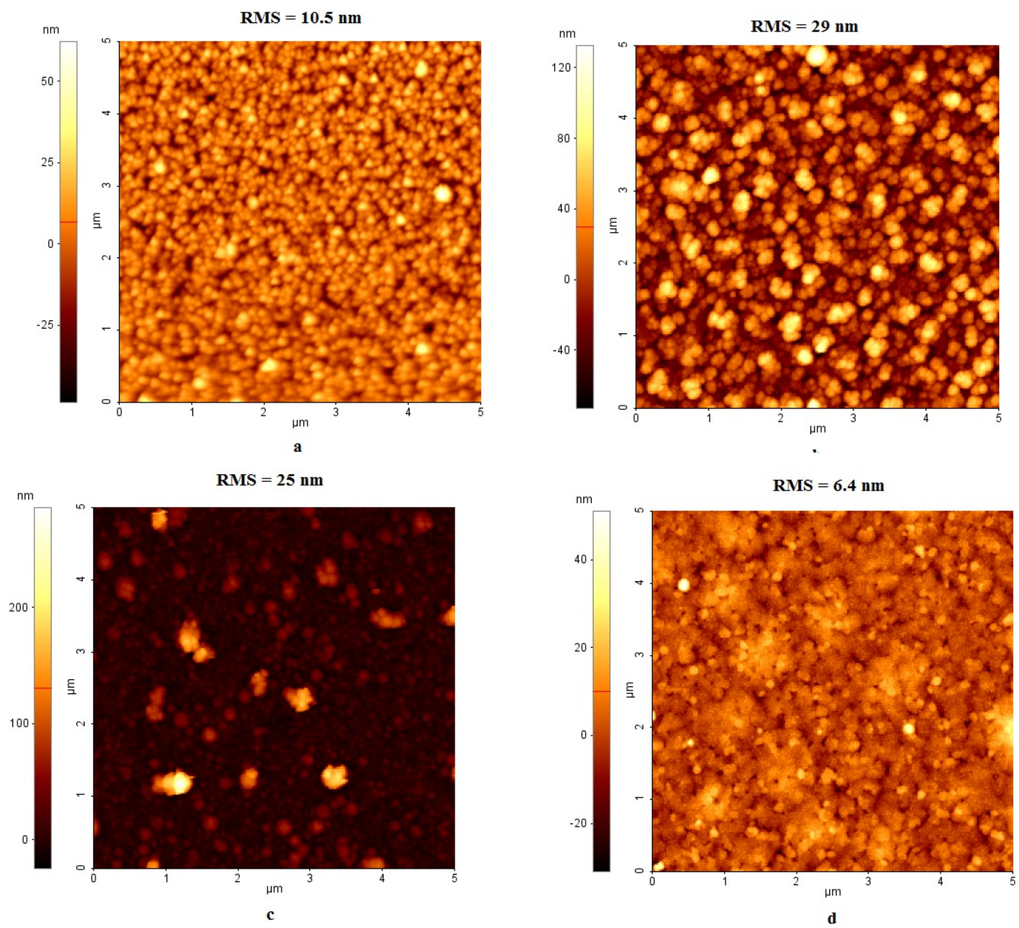


Fig. 1. AFM images for SBN:50 thin films deposited at different thermal contact conditions of substrates: *a* - SBN:50/Pt:Si (111) ; *b, c, d* - SBN:50/MgO (001)

For SBN:50 thin films deposited on Pt:Si (111) and MgO (001) substrates with high thermal contact (Fig. 1 - a, d), the amount of large grains is lower than in the case of MgO substrates with unsecured thermal contact (Fig. 1 - b, c). The roughness evaluated by RMS (Root Mean Square) shows a smaller value (RMS = 6.4 nm) for SBN:50 thin film grown on a MgO (001) substrate with high thermal contact than the samples with unsecured thermal contact of substrates (RMS = 25 and 29 nm).

3.2. Structural investigations

XRD spectra recorded on thin films deposited on Pt:Si (111) and MgO (001) substrates indicates that the *c* axis-oriented SBN:50 thin films are presented for all substrates. For the layers deposited on a Pt:Si substrate with secure thermal contact, peaks coming from secondary phases are evidenced. The 2theta-omega thin film pattern with a 3^{o} omega offset superimposed with the SBN:50 ceramic target spectrum is presented in figure 2.

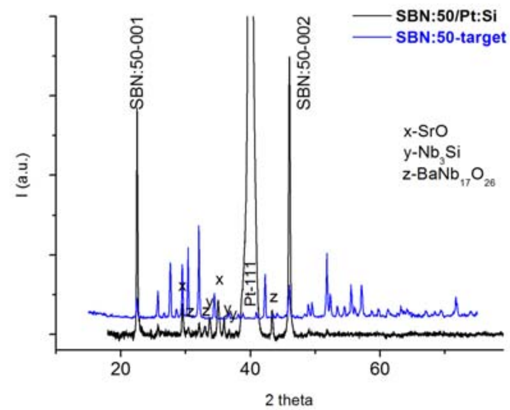
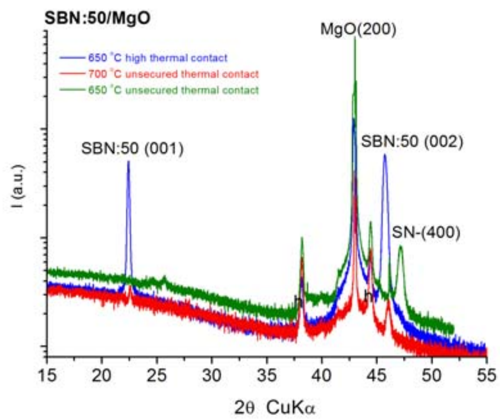


Fig. 2. The XRD 2theta-omega scan with an omega offset of 3^{o} of SBN:50 thin film deposited on a Pt:Si (111) substrate with secured thermal contact

It can be noticed the relative intense (00l) peaks of the *c*-axis oriented SBN:50 phase, small peaks of a polycrystalline SBN:50 phase and some extra peaks which could presumably assigned to SrO (JCPDS 48-1477), $BaNb_{11}O_{26}$ (JCPDS 45-0146) and Nb_3Si (JCPDS 34-1043) phases. In figure 3 are presented the XRD spectra for the SBN:50 thin films deposited on MgO (001) substrates with secured and unsecured thermal contact. It can be observed that the layer composition and texture is highly sensitive to the thermal contact conditions between the substrate and the heater. Highly crystalline and highly *c* axis-oriented SBN:50 thin films are obtained when the substrate has a secure thermal contact with the heater. The *c*-parameter value for the SBN:50 phase is 3.965 Å. With no secure thermal contact, the growth of the SBN layered is inhibited. At lower temperature (650°C) and unsecured thermal contact of substrate, a $SrNb_2O_6$ (SN) (JCPDS 45-02



At higher temperature (700°C) an extremely poor crystallized SBN:50 phase could be detected, as it can be seen from the figure 4 as well, where the regions of (001) and (002) SBN:50 peaks are presented in details.

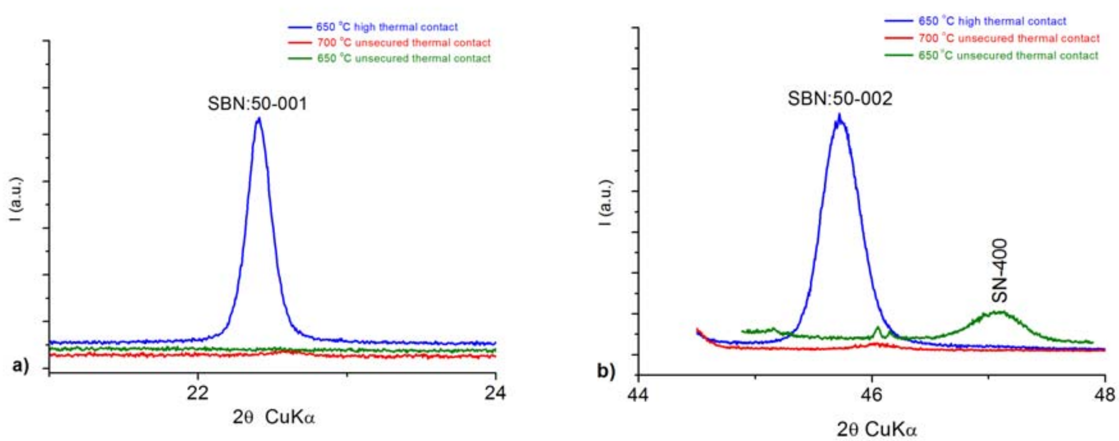


Fig. 4. (001) (a) and (002) (b) peaks of SBN:50 thin films deposited on MgO substrates with secured and unsecured thermal contact

4. Conclusion

SBN:50 thin films were deposited on Pt:Si (111) and MgO (001) substrates by RF-PLD technique. The influence of the substrate thermal contact conditions on the growth of SBN thin films was investigated. X-ray diffraction spectra performed on the SBN:50/Pt:Si thin film grown in secure thermal contact conditions has shown that the layers are predominantly polycrystalline, with a relatively *c*-axis texture, but also with some secondary phases. For SBN:50/MgO thin film obtained with a secure thermal contact between the substrate and the heater, AFM results show uniform nanostructured morphologies, with smaller amount of large grains compared to the samples deposited on MgO substrates with unsecure thermal contact. Moreover, the roughness (RMS = 25 - 29 nm) is higher for layers grown on substrates in unsecure thermal contact regime. XRD analyses evidenced single-phase, highly *c*-axis oriented SBN:50/MgO thin film when a secure thermal contact between the substrate and the heater during deposition is assured.

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