Nuclear investigations on ferroelectric materials for high frequency applications

Proposal submitted to the Program Advisory Committee (PAC) for the "Campaign of experiments 2017"

Dr. ing. Marian Gabriel BANCIU,
National Institute of Materials Physics (INCDFM)
105 bis Atomistilor St.
Magurele 077125, Jud. Ilfov
Tel. 021-2418123; Fax. 021-03690177
Mobile: 0744174400
E-mail: gbanciu@infim.ro

Dr. Dan PANTELICA
"Horia Hulubei" National Institute of Nuclear Physics (NIPNE)
Phone: 021 404 2332
E-mail: pantel@nipne.ro

1. Scientific motivation of the proposed work

1.1. Investigations on BNT-$BT_{0.08}$ films

Recent research showed that the (1-*x*)($Bi_{0.5}Na_{0.5}$)TiO_3-*x*$BaTiO_3$ ferroelectric films (for short BNT-$BT_{0.08}$) exhibit attractive features, which recommend them for microwave agile devices [1]. Interesting properties were observed for *x* taking values in the range 0.06 – 0.1. The ferroelectric tunable capacitors are an interesting alternative to the conventional varactors, which show a very low power handling capability.

The BNT-BT material was manufactured by using the sol-gel technique and spin coating on MgO single-crystal substrates. After a 20 s coating at 3000 rotations per minute. The solvent was evaporated by warming up the samples for two minutes at 200 ºC. A further heating at 400 ºC for four minutes was used to eliminate the organic components. The deposition and heating cycle was repeated for several (usually five) times. In order to achieve the full crystallization, the samples are annealed in oxygen at 800ºC for 15min. [1]

Layers with thickness between 350 and 550 nm were achieved by changing the number of cycles of coating [1]. As top electrode needed by the IDC (interdigital capacitor) or CPW (coplanar wave guide) devices, a ~200nm thick Au layer was deposited. For other devices such as MIM (metal insulator metal) capacitor, a bottom electrode of Ir was deposited.

The inset in Fig. 1 show the tunability of the BNT-BT films. For *x*=0.08, cavity measurements working in $TE_{01\delta}$ mode at 12.5GHz obtained a dielectric constant of 204 and a dielectric loss tangent of 0.15. In addition, the MIM device working at 2.45 GHz showed a 50% tenability for a 526 kV/cm [1].

The elemental analysis is very important for the BNT-$BT_{0.08}$ films. On one side, the RBS measurements will give information about the content *x* of barium and content (1-*x*) of bismuth. On the other side, such light elements as sodium or oxygen can be investigated by ERDA experiments.

1.2. Investigations on BST films and single crystals

The (Ba_{1-x}, Sr_x)TiO_3 material is another ferroelectric material with high potential for high frequency devices. The tunability curves for a IDC structure on a BST film are shown in Fig. 2 [2-3]. In order to benefit of the tunability and to avoid very high bias voltage, thin films of barium irontium titanate can be deposited on different substrates and investigated. The deposition methods can vary from RF magnetron deposition, pulsed laser deposition, pulsed electron beam deposition etc. However, not all deposition methods preserve the target stoichiometry. For example, special problems occur when RF magnetron deposition is used. Therefore, advanced methods able to provide compositional information of the deposited films need to be employed. Among these methods, the Rutherford Backscaterring (RBS) method is a very powerful experimental technique for thin film analysis. Moreover, the RBS method can provide also information on the film thickness, which is very important for an appropriate estimation of dielectric parameter in THz domain. The THz investigations will be carried out with the Time Domain THz spectrometer in the National Institute of Materials Physics.

Most of the published works are focused on BST with strontium content x = 0.4 or 0.5 leading to a Curie point close to the room temperature. However the research will consider a variety of Sr content, because interesting applicative results can be achieved in both ferroelectric phase (small x) and paraelectric phase (high x).

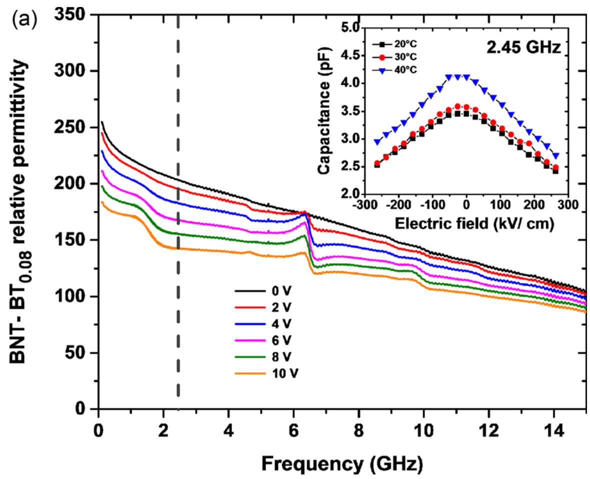

Fig. 1. Frequency dependence of the dielectric permittivity of the BNT-BT film measured using a metal-insulator-metal device. Inset: capacitance variance with voltage at 2.45GHz [1].

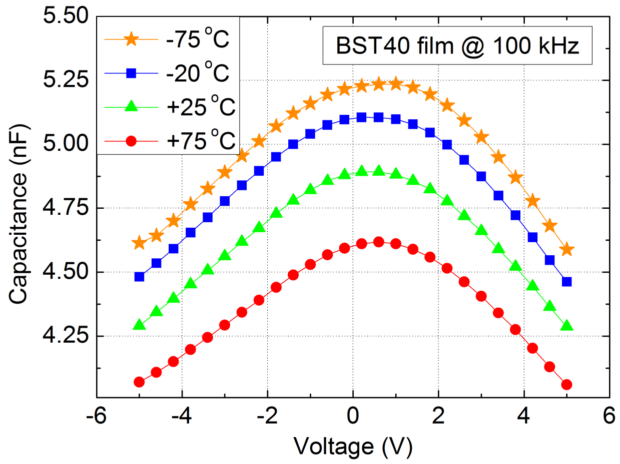

Fig. 2. Tunability of a barium strontium titanate thin film with x=0.4 strontium content, measured in previous research [3].

Usually, the deposited films are ~200 nm thick. However, in order to investigate the ferroelectric films in Terahertz domain, 2 μm thick films were deposited on MgO and intrinsic Si substrates.

The barium strontium titanate (Ba_{1-x}, Sr_x)TiO_3 is a very interesting ferroelectric material with great potential for applications to high frequency electronics.

Our group in National Institute of materials Physics has a significant experience in developing bulk and thin film barium strontium titanate [2-5]. Recently, as a result of a collaborative work between the National Institute of Materials Physics and the National Institute of Lasers, Plasma and Radiations, single crystals of(Ba_{1-x}, Sr_x)TiO_3 with x=0.1 and x=0.2 were grown (see Fig 3).

Fig. 3. (Ba_{1-x}, Sr_x)TiO_3 crystals x=0.1 - 0.2.

Preliminary XRD investigations showed some twining in the grown crystals of barium strontium titanate. We intend to investigate the crystalline orientation by using Rutherford backscattering spectrometry in channeling geometry (RBS-C). The experiment will be performed using the precision goniometer installed in IBA reaction chamber at the 3MV Tandetron. We intend to perform also RBS investigations to measure the depth profiles of Ba, Ti and Sr. The RBS investigations will be very useful for obtaining the depth profile of Ba, Ti and Sr elements in the samples. Moreover, in order to investigate the oxygen profile in the samples, experiments of elastic recoil detection analysis (ERDA) will be carried out at the 9MV Tandetron by using ^{63}Cu ions of 80 MeV. In addition, non-Rutherford backscattering (NRBS) experiments will be conducted in order to analyze the oxygen in the samples.

Due to the fact that the BST crystals were grown in N_2 athmosphere, the BST crystals exhibit oxygen deficiency (not detectable in X-ray diffraction) and the dielectric and optical properties are to be improved. Further thermal treatment is required for crystals in order to correct that oxygen deficiency. The ERDA experiments will offer useful information about the oxygen content.

1.2. Investigations on thin films

The advantages of the RBS method [6] :

- Quantitative elemental analysis without standards due to the high cross section, which is possible to be calculated analytically
- Excellent sensitivity (better than 0.01 monolayer) for heavy elements in a light substrate due to the smooth dependence of yield on the square of atomic number
- Possibility of profiling elements and investigation of interfaces or up to 1000 nm thick multi-layer structures; the RBS measurements will allow the investigation of the compositional gradient.
- Possibility of gathering information on topography, surface relaxation, epitaxial growth for single-crystals
- Possibility of location of substitutional or interstitial impurities and damage and annealing investigation
- Non-Rutherford cross-sections for light element profiling for enhanced sensitivity in light elements

2. Implementation of the experiments
2.1. Investigations at 3MV Tandetron Accelerator

2.1.1. RBS experiment on $(1-x)(Bi_{0.5}Na_{0.5})TiO_3$-$x BaTiO_3$ deposited on MgO (100) and Si substrates ~ 12 samples **3days**

2.1.2 RBS experiments on $(Ba_{1-x}, Sr_x)TiO_3$ (x=0.1 and 0.2) films and crystalline samples **3days**

2.2. Investigations at 9MV Tandetron Accelerator

2.2.1. ERDA experiments using 80MeV in order to investigate the oxygen concentration profile for some $(Ba_{1-x}, Sr_x)TiO_3$ crystalline and thin film samples and for BNT-BY films **4 days**

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