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Review

The preparation and refractive index of BST thin films

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Abstract

Radio-frequency magnetron sputtering technique is used to deposit $Ba_{0.65}Sr_{0.35}TiO_3$ (BST) thin films on fused quartz substrates. In order to prepare the high-quality BST thin films, the crystallization and microstructure of the films were characterized by X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM) and atomic force microscopy (AFM). More intense characteristic diffraction peaks and better crystallization can be observed in BST thin films deposited at 600 °C and subsequently annealed at 700 °C. The refractive index of the films is determined from the measured transmission spectra. The dependences of the refractive index on the deposition parameters of BST thin films are different. The refractive index of the films increases with the substrate temperature. At lower sputtering pressure, the refractive index increases from 1.797 to 2.197 with pressure increase. However, when the pressure increases up to 3.9 Pa, the refractive index reduces to 1.86. The oxygen to argon ratio also plays an important effect on the refractive index of the films. It has been found that the refractive index increases with increase in the ratio of oxygen to argon. The refractive index of BST thin films is strongly dependent on the annealing temperature, which also increases as the annealing temperature ascends. In a word, the refractive index of BST thin films is finally affected by the films' microstructure and texture.

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Keywords: BST thin film; RF magnetron sputtering; Deposition parameter; Refractive index

Contents

1. Introduction

Ferroelectric thin films are very promising for a wide range of applications such as uncooled infrared detectors and focal plane arrays [1], non-volatile memories with low

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switching voltage [2], planar waveguides [3] and electro-optic devices [4]. Among these materials, $Ba_{1-x}Sr_xTiO_3$(BST) thin films have attracted much attention because of its unique combination of large dielectric constant, high refractive index, large electro-optic coefficient and low optical losses [5–9].

There have been several reports on the electrical and ultraviolet–visible optical properties of $Ba_{1-x}Sr_xTiO_3$ thin films [10–14]. Recently, Roy et al. [15] reported that the optical property in sol–gel was derived from $Ba_{0.5}Sr_{0.5}TiO_3$ thin films using a 0.1 M precursor solution. Xu et al. [16] reported the optical properties of $Ba_{0.7}Sr_{0.3}TiO_3$ thin films. However, no more detailed refractive index of $Ba_{1-x}Sr_x TiO_3$ thin films has been provided. So far, most studies in ferroelectric thin films have focused on the general optical properties, and, to our knowledge, there are no comprehensive reports concentrating on the refractive index of BST thin films varying with the deposition parameters, especially the sputtering pressure and the ratio of argon to oxygen.

The object of this paper is to prepare high-quality BST thin films and obtain the relationship between the refractive index of BST thin films and the deposition parameters (the substrate temperature, the sputtering pressure, the ratio of argon to oxygen and the annealing temperature). It is believed that the dependence of the refractive index of BST thin films on the substrate temperature, the sputtering pressure, the ratio of argon to oxygen and the annealing temperature should be important to optimize the design of electro-optic devices to improve the potential performance of ferroelectric thin-film electro-optic devices.

2. Experimental

Radio frequency (RF) magnetron sputter techniques have been used to deposit BST thin films on fused quartz. The BST ceramic target was prepared from high-purity $BaCO_3$, $SrCO_3$ and TiO_2 powders (purity 99.9%) using standard solid-state process. The powders were mixed in the planetary milling for 4 h using a plastic container with agate balls. Disk-shaped specimens of 70 mm in diameter and 3 mm in thickness were obtained by the uniaxial pressing at 100 MPa. The disk-like BST is sintered at 1100 °C for 2 h and at 1350 °C for 2 h in order to densify the target.

The substrates were rinsed in acetone, ethanol and distilled water with a supersonic wave apparatus, and dried by nitrogen gas sequentially before being loaded into the growth chamber.

With a fixed target-to-substrate distance, the sputtering was carried out in an Ar and O_2 (99.99%) atmosphere by supplying 120 W RF power at a frequency of 13.56 MHz. A cryopump coupled with a rotary pump was used to achieve 1×10^{-5} Pa pressure before introducing Ar and O_2. A mixture of oxygen and argon at various mixing ratios ranged from 1:1 to 1:4 with a total flow of 24 sccm. During the sputtering, the substrate temperatures were kept at room temperature (RT), 560, 600 or 650 °C. The temperature was controlled using a feedback-controlled heater. The films were cooled to room temperature before removing them from the deposition system oxygen at a temperature of 650, 700, 750 or 800 °C for 2 h in a tube furnace. Post-annealing in oxygen was done to reduce any oxygen deficiency in the films.

Crystalline phases of the BST samples were identified by X-ray diffraction (XRD, Rigakn D/MAX-IIIC). The XRD operating parameters included a Cu Kα of wavelength $\lambda = 0.154176$ nm, scan speed of 5°/min and scan range of 20–67°. The surface microstructure and cross-sectional morphology were observed by field emission scanning electron microscopy (FESEM, JOEL JSM 6700F). The surface morphology was examined by atomic force microscopy (AFM; Digital Instruments, NanoScope IIIA). The optical transmission spectrum of BST films were measured in the wavelength range of 190–900 nm using a double beam ultraviolet–visible spectrophotometer (Shimadzu UV-1601 UV/VIS). The envelope method was used to calculate the optical constants.

3. Results and discussion

3.1. The preparation

The crystallinity of BST films deposited on fused quartz substrate was investigated by XRD spectrum analysis as shown in Fig. 1. The as-deposited film and the film for growth at lower substrate temperature (curve 1(a) and curve 1(b)) are found to be a mixed amorphous/polycrystalline phase. The diffraction peaks are not sharp, and the crystallinity is not perfect.

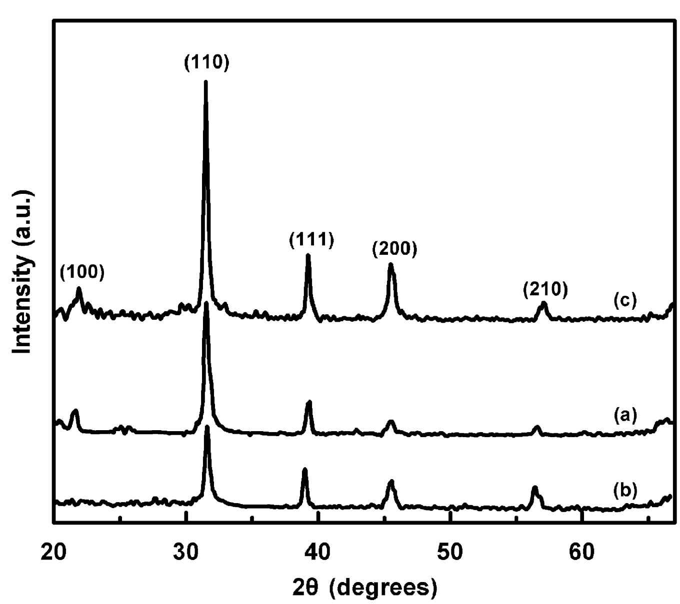

Fig. 1. The XRD patterns of BST thin films. (a) Deposited at 600 °C without annealing, (b) deposited at 560 °C and subsequently annealed at 700 °C, (c) deposited at 600 °C and subsequently annealed at 700 °C.