Journal of Alloys and Compounds 687 (2016) 458–462

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Effect of electric field on the dielectric properties of the Barium Strontium Titanate film

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ARTICLE INFO

Article history:
Received 25 January 2016
Received in revised form
24 May 2016
Accepted 29 May 2016
Available online 30 May 2016

Keywords:
$BaSrTiO_3$ film
Dielectric characterization
THz
Electrically tunability

ABSTRACT

We investigated the tunability of the dielectric characterization of the Ferroelectric Barium Strontium Titanate ($Ba_{0.5}Sr_{0.5}TiO_3$) thin film on the Strontium Titanate substrate by a dc bias electric field through the terahertz time-domain spectroscopy. The tunability of the real part of the permittivity reached up to 74.8% and the imaginary part of the permittivity reached up to 33.6% when the bias electric field up to 30 V. And the results are attributed to the soft mode hardening caused by the electric field.

1. Introduction

Due to the excellent nonlinear optical properties, ferroelectric and electro-optical physical properties and comprehensive application in various areas, ferroelectric materials have been drawn great attention both in scientific research and technology development [1]. The ferroelectric perovskite oxide materials Barium Strontium Titanate ($Ba_xSr_{1-x}TiO_3$) has attracted great interests for several decades due to its low loss and composition dependent Curie temperature (Tc) [2], which ranges from −163 to 120 °C as x could varied from 0 to 1 continuously. Now $Ba_{1-x}Sr_xTiO_3$ has been widely used in the design of electronic devices intended for controlling radiation in the optical and microwave frequency ranges [3], and used in the dynamic random access memories [4,5] and the functional elements in devices [6,7].

However for the bulk BST material: the annealing temperature demands very high to obtain the good crystallinity; the size is too large and is not convenient to do device integration; and the dielectric constant is so big that demands a very complicated impedance matching circuit [2], all these characters hamper the bulk BST in the integrated applications. Thin BST film offers advantages over bulk BST for tunable applications. Large electric fields (0–400 kV/cm) can be achieved in thin film BST (224 nm) using low voltages [2]. For ferroelectric BST film, the tunable properties are most researched in the microwave region and in the terahertz range by the means of dc electric-field [8–10], or by the means of codoping and adjusting sintering temperature for attain the high modulation of $Ba_{0.67}Sr_{0.33}TiO_3$ ceramics [2], or adopted the method of temperature control [2]. Due to the excellent optical performance, many properties of the film have been experimentally and theoretically studied [11]. And for the investigation of modulate properties of the thin film BST, many solutions have been proposed in literature [8,12].

Recently, there are some observations of the BST film tunability have been reported [13–15], but most of these literature focus on the temperature dependent photoelectric properties. It is necessary for the permittivity to be modulated over a range as wide as possible around room temperature for practical applications. In this manuscript, we report a study of the dielectric THz properties and the electrical-field-induced tunability of BST film at room temperature.

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http://dx.doi.org/10.1016/j.jallcom.2016.05.324

2. Experiment

The BST film samples were prepared from a stoichiometric $(Ba_{0.5}Sr_{0.5})TiO_3$ target on the single crystal STO(100) under the conditions listed in Table 1. The substrates were ultrasonically cleaned with acetone and isopropanol prior to BST deposition. Films prepared at the condition of Ar/O_2 is 40/10. After the deposition, the film was immediately annealed for 10 min in a vacuum condition inside the quick anneal oven, and then cooled to room temperature by nitrogen. The structural analysis of the film has been performed by XRD (X-ray Diffraction) scan and shown in the Fig. 1. Mainly STO (100) and STO (200) peaks are observed, which clearly shows that highly oriented the film prepared on STO (100) substrate. As could be seen that the film is cubic crystal. The calculated lattice constants is 3.965 nm, these values are very closed to those of bulk samples [16]. The surface roughness of the BST films analyzed by atomic force microscopy (AFM), and the results is shown in Fig. 2. The root mean square (RMS) roughness of BST was 2.038 nm, indicating sufficiently smooth surfaces for dielectric measurements with THz-TDS and IDEs are realized. The dimensions of the studied substrates were 2 mm × 2 mm × 0.5 mm. We measured the thickness of the film of 25 nm (4% accuracy) by Step profiler.

The gold electrodes were prepared by standard photolithography and ion beam etching of a 20 nm thick titanium/200 nm thick gold film deposited on top of the BST film. The interdigitated capacitor structure of 16 mm × 10 mm size was composed of 5 μm wide gold lines separated by 15 μm wide gaps, as shown in Fig. 3.

We used a THz time domain spectrum (THz-TDS) system produced by the Zomega Terahertz Corporation of the US to measure the transmittance spectrum at room temperature. The fiber femtosecond laser beam was divided into two beams (named the pump beam and the probe beam, respectively) by a polarized beam splitter. The THz pulses transmitted through the sample were detected using a usual electro-optic sampling scheme with another 1 mm thick ZnTe (110) crystal and a pair of balanced Si photodiodes. The TDS probes the in-plane response of the sample. The measured frequency resolution is 10 GHz.

3. Results and discussion

The measurements were carried out in two steps [8]: firstly, measurement of a reference wave form *Eref*(ω,V) with a bare substrate STO and the result was shown in Fig. 4(a); secondly, measurement of a signal wave form *Es* (ω, V) with different bias electric field with the thin film on a substrate as shown in Fig. 4(b), where the V is the bias external field. The transmission is extracted from the ratio of the Fourier-transformed amplitude spectra of the signal and reference, defined as:

$$T(\omega,V)=\frac{E_S(\omega,V)}{E_{ref}(\omega,V)} \tag{1}$$

As can be seen, the signal waveforms transmitted through the BST film show little time delay with increasing bias field, as shows in Fig. 4(b), the transmittance waveform shifts about 0.35 ps when the bias electric field up to 30 V, compared to the waveform without bias field. And that the transmitted THz signal decreases with the increasing electric field.

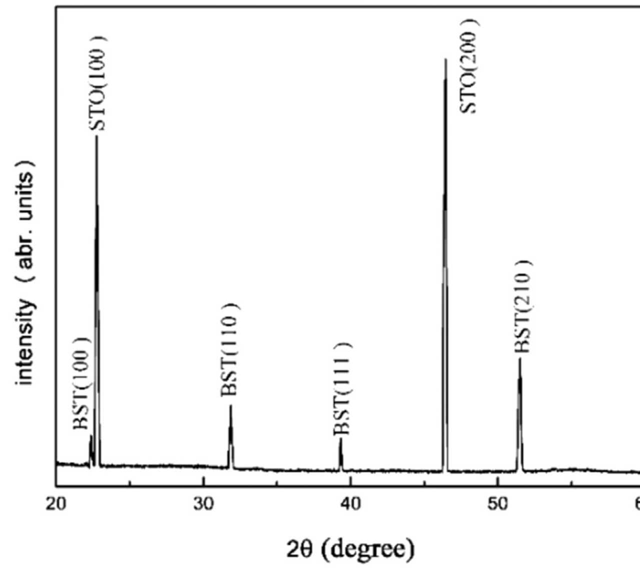

Fig. 1. XRD patterns for BST thin film on the substrate of STO. Diffraction peaks from a STO (100) substrate are marked in the figure.

Both the real parts and imaginary parts of the dielectric constant of the BST thin film have been evaluated by numerically inverting the expression for $T(\omega)$ [17]:

$$T(\omega)=\frac{2N_f(N_s+1)\exp\left[i\omega\left(N_f-1\right)d_f/c\right]}{\left(N_f+1\right)\left(N_f+N_s\right)+\left(1-N_f\right)\left(N_f-N_s\right)\exp\left[i\omega\left(N_f d_f/c\right)\right]} \tag{2}$$

where, d_f is the film thickness, c is the speed of light and $N_s=\sqrt{\varepsilon(\omega)}$ is the complex optical refractive index of the substrate as a function of ω. The technique allows us to determine the dielectric response of the thin film in the range from 0.2 THz to 1.2 THz [17]: $\varepsilon(\omega)=\varepsilon'(\omega)+i\varepsilon''(\omega)$.

Fig. 5 shows the complex dielectric spectra of the BST film under different values of the bias electric field. The detectable frequency range is from 0.2 THz to 1.2 THz. It is clearly seen in Fig. 5 that the electric-field-induced changes are significantly weaker when below 25 V. The both real parts ε' and the imaginary parts ε'' were demonstrated in Fig. 5, which showed both the real part and the imag part of the permittivity decrease with an increasing bias electric field. As can be seen from Fig. 5(a), the value of the permittivity is about 6000, the values of the calculated are larger than the previous results [3], which is consistent with the other report [18], the calculated dielectric constant are inversely proportional to the thickness of the film.

Fig. 6 shows the electric field tunability of the complex permittivity from 0.2 THz to 1.2 THz. From the figures, we could get the tunability of the dielectric constant for different bias field using the following equations:

Table 1
Sputtering conditions for BST deposition.

Specimen	Conditions					
	Target diameter	Source to substrate distance	RF power	Sputtering gas	Substrate temperature	Gas pressure
BST film	3 inch	4 inch	140 W	Ar/O_2	400 °C	50 mT