Optical Materials 60 (2016) 383–386

Contents lists available at ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

Optical-induced absorption tunability of Barium Strontium Titanate film

Chunya Luo [a, d], Jie Ji [a], Jin Yue [a], Yunkun Rao [b], Gang Yao [a], Dan Li [d], Ying Zeng [b], Renkui Li [b], Longsheng Xiao [d], Xinxing Liu [a], Jianquan Yao [a, c, d], Furi Ling [b, *]

[a] Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, Wuhan 430074, China
[b] School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China
[c] College of Precision Instrument and Opto-electronics Engineering, Tianjin University, Tianjin 300072, China
[d] Institute of Information Science and Technology, Department of Physics and Mechanical & Electrical Engineering, Hubei University of Education, Wuhan 430205, China

A R T I C L E I N F O

Article history:
Received 1 February 2016
Received in revised form
18 August 2016
Accepted 21 August 2016

Keywords:
Optical tunability
Absorption response
$BaSrTiO_3$
THz

A B S T R A C T

The absorption tunability of 100 nm thickness of ferroelectric Barium Strontium Titanate ($Ba_{0.5}Sr_{0.5}TiO_3$) thin films with different densities of pumped optical field is measured by terahertz time-domain spectroscopy in the range of 0.2 THz – 1.2 THz at 19 °C. Experimental results show that the absorption coefficient of BST film is approximately at 5000 cm^{-1}–20000 cm^{-1} in the range of 0.2 THz – 1.2 THz and the absorption coefficient reached up to 16% when we applied the optical field up to 600 mW. The theoretical calculations reveal that increasing photoexcitation fluences is responsible for the increasing of transmission change in the conduction current density cause the absorption coefficient varied.

1. Introduction

For the sake of comprehensive application prospects in various areas such as optical, electric, and microwave devices in wireless communications and control systems, and possessing the excellent nonlinear optical properties, ferroelectric and electro-optical physical properties, ferroelectric materials have drew great attention both in scientific research and technology development [1]. In which, the ferroelectric materials Barium Strontium Titanate ($Ba_xSr_{1-x}TiO_3$, abbreviated as BST) has been researched for several decades. BST materials has widely used in the microwave tunable devices and dynamic random access memories [2,3]. BST materials are promising candidates for use in the design of electronic devices intended for controlling radiation in the optical and microwave frequency ranges [4]. $Ba_{1-x}Sr_xTiO_3$ is currently a material for microwave applications due to its low loss and composition dependent Curie temperature (Tc) [5], which ranges from −163 to 120 °C for x = 0 to x = 1 respectively. We chose that the Ba/Sr ratio is 1 to realize better tunability, for in that condition, the Tc is just at around room temperature, which is more favorable for practical device application. In order to make up the defects of the bulk BST, such as the sintering temperature demands very high to obtain good crystallinity, the dielectric constant is too high to inevitably leads to a very complicated impedance matching circuit [5], we prefer to do the research on the BST film.

Thin film BST 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 [5]. And on the modulate properties of the thin film BST, many methods have been proposed in literature. The agility of the modulate properties comes from the modification of temperature [6] or external dc (low-frequency) electric field [7]. The generalization of terahertz (THz) communicating systems in the objects of space program and military contributes significantly to the development of miniature and agile modulators. The use of a single reconfigurable modulator allows tuning its operating frequency to different communications standards and so, permits replacing multiband elements in standard systems. Ferroelectric materials, which are characterized by a high permittivity up to the THz range [7] offer a possibility of its tuning by means of external field. Whereas, these tunable properties are most researched in the microwave region [8,9] or in the terahertz

* Corresponding author.
E-mail address: lingfuri@mail.hust.edu.cn (F. Ling).

http://dx.doi.org/10.1016/j.optmat.2016.08.012
0925-3467/

range but by the means of dc electric-field [7–9], or by the means of codoping and adjusting sintering temperature for attain the high modulation of Ba0.67Sr0.33TiO3 ceramics [5], or adopted the method of temperature control [5]. One of the works where STO shows large change in the complex refractive index with temperature [10], although the modulation capability is superior to the pumped optical tunability, the thermal tunability was not so accurate. Due to the excellent optical performance, many properties of the film have been experimentally and theoretically studied [11].

In his work, the transmission spectrum of BST film in the THz range with a varied external pumped optical field is measured. The absorption coefficient of the BST thin film at THz frequencies was derived from the measurement data. The pumped optical field dependence of absorption coefficient of the BST thin film thus evaluated at THz frequencies.

2. Experiments

The BST film samples prepared via a standard magnetron sputtering method. The films were sputtered from a stoichiometric $(Ba_{0.5}Sr_{0.5})TiO_3$ target onto TiO_2 (100 nm)/SiO_2 (100 nm)/Si substrates. And then the step profiler reveals a thickness of 104 nm (4% accuracy). The dimensions of the studied substrates were 2 mm × 2 mm × 0.5 mm.

The THz time domain spectrum (THz-TDS) system produced by the Zomega Terahertz Corporation of the US was used to measure the transmittance spectrum at 19 °C, which the signal to noise ratio of the terahertz data/measurements was above the 50 dB. Fig. 1 shows that in the fiber femtosecond laser beam was divided into two beams (named the pump light 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 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, and the spot size is 3 mm in diameter. An all-solid-state continue-wave (cw) green laser (center wavelength 532 nm) was employed in the experiment to provide external optical pumping. The light is obliquely incident upon the surfaces of the samples at an angle of 45°and the spot size is 5 mm in diameter [12]. To make sure that the sample restored to its previous dynamic behavior, we performed a thermal process after each measurement. The sample was placed in a furnace at 220 °C for 3 h, and no external field was applied to it in this recovery process.

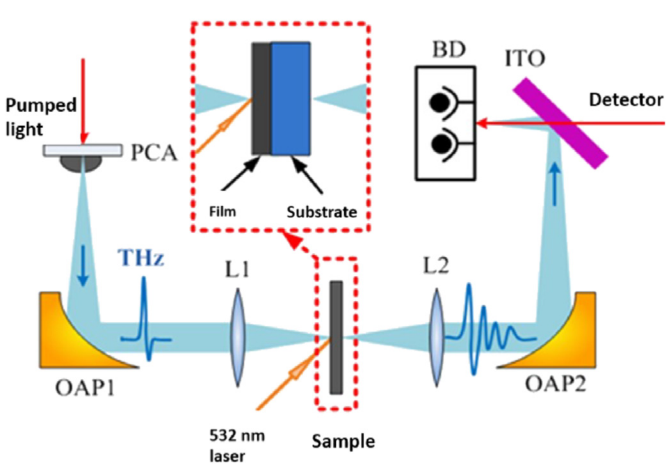

Fig. 1. Installation diagram of the TDS. A green laser is obliquely incident upon the surface of the sample at an angle of 45°with respect to the polar axis.

In the experiments the sample was casted with the different external optical densities fields onto the BST film. A 532 nm green laser was employed in the experiment to provide external optical pumping and the optical density was increased to 600 mW. We calculated the complex transmittances and the intrinsic phases in frequency domain from the waveforms in Fig. 2b through a Fourier transform. The experiments was carried out in two steps [7]: firstly, measurement of a signal wave form *Es* with the thin film on a substrate in the path of the THz beam; and secondly, measurement of a reference wave form *Eref* with a bare substrate see Fig. 2a.

3. Results and discussion

Fig. 2a demonstrates the reference wave form *Eref* with the substrate and Fig. 2b shows the signal results of the zero-field (0 mW) wave form and the difference wave form of 200, 400 and 600 mW optical density applied. As can be seen that these signals almost have the same shape except little change with increasing optical field amplitude, as shows in Fig. 2a that the transmittance waveform shifts about 0.025 ps when the pumped optical field power is 600 mW, compared to the without pumped optical field, and that the transmitted THz signal increases when the pumped optical field increasing.

We calculate the transmission spectrum and the intrinsic phase shift of the sample by the Fourier-transform method:$T(\omega) = \frac{E_S(\omega)}{E_{ref}(\omega)}$.

We have calculated the complex refractive index spectrum of the BST film in the research. Taking account of the multiple reflections inside the Si substrate, both the real and imaginary parts of the refraction index of the BST thin film have been evaluated from the measured waveforms using Fresnel's equations [13].The technique allows us to determine the refractive index of the thin film in the range from 0.2 THz to 1.2 THz [14]: $N_f(\omega) = n(\omega) + ik(\omega)$. In which, the $n(\omega)$ and the $k(\omega)$ are the real and imaginary parts of the refractive index of the film, respectively. The complex refractive index N_f was then retrieved by numerically inverting the expression for $T(\omega)$ [14], from the THz transmittance spectra by formula (1).

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

Here, d_f is the film thickness, c is the speed of light and N_s is the complex optical refractive index of the substrate as a function of ω.And in order to investigate the influence of the pumped optical field on the refractive index of BST film, we calculate the frequency dependence of the real and imaginary part of the complex refractive index as shown in Fig. 3a and b. Fig. 3 demonstrates the optical properties of the BST film at the different external optical fields. The detectable frequency range is from 0.2 THz to 1.2 THz. The value of the real part of the refraction index is 35–50, and the imaginary part of the refraction index 10–40. It is showed that the n demonstrated a decreasing with the optical density increased and the k showed an inversely tendency, which shows a certain tunability of the optical property.

In order to investigate the absorption properties of the BST film, the frequency dependence of absorption coefficient was calculated with the Eq. (2) [15], as shown in Fig. 4a.

$$k(\omega) = \alpha(\omega)c/2\omega \quad (2)$$

As can be seen from Fig. 4a it is found that the absorption coefficient is about 15000 in the range and the absorption coefficient is decreasing with the frequency increased. For evaluate the