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Optically tuned dielectric property of barium titanate thin film by THz spectroscopy

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ABSTRACT

The dielectric property of ferroelectric barium titanate ($BaTiO_3$) thin film with optical field was investigated by terahertz time-domain spectroscopy at room temperature. Experimental results showed that dielectric constant of BTO film was increased with the optical pump powers, and tunability of the real part of dielectric constant could be reached to74%. The reason of realizing high modulation depth could be explained as photorefractive and photothermal effects. Furthermore, the variation of refractive index displayed a monotonically increase with the optical powers.

1. Introduction

Optical or electrical control of the propagation of terahertz (THz) radiation has great importance in the current technology, and functional devices in the THz range have recently received considerable attention [1–3]. Ferroelectric materials play an important role in the research of functional devices operating in the THz range because of their response time, dielectric loss and tunability [4]. Ferroelectric thin film offers advantages over ferroelectric bulks for tunable applications [5]. Large electric fields (0–400 kV/cm) could be achieved in thin film with thickness of nanometer level by using low voltages [4]. Among the ferroelectric perovskite, barium titanate ($BaTiO_3$, BTO) has received most theoretical attention, and may be regarded as prototypical materials. As a function of ascending temperature, both materials undergo rhombohedral to orthorhombic, to tetragonal and to cubic transitions, with all non-cubic phases being ferroelectric [6,7].

$BaTiO_3$ is the most important lead-free material with excellent dielectric and ferroelectric properties for wide applications, including capacitors, memories and so on [8–12]. To improve the device performance and meet the continuing demand, tremendous efforts have been pursued to modify the dielectric properties of $BaTiO_3$-based materials, such as substitution and introducing "interface structure" [13–15]. Lee et al. enhanced the dielectric property of ferroelectric thin film by preparing ferroelectric $BaTiO_3$/$SrTiO_3$ superlattice and introducing the strains in the ferroelectrics [16]. Zhu et al. found that dielectric properties of $BaTiO_3$ ceramics could be modified by Ca-substitution [12].

As it is known, the polarization in ferroelectrics (in contrast to regular dielectrics) is an essentially nonlinear function of the electric field and temperature field, most studies on the dielectric tunability of ferroelectric thin films have been conducted with external electric and temperature fields [17–21]. In this letter, we investigate the dielectric property of BTO thin film with different external optical pump powers at room temperature (approximately 291 K) in the THz range.

2. Experimental details

A radio frequency magnetron sputtering apparatus was employed to prepare BTO thin film. The target were single-crystal BTO, which was (100)-oriented and purchased from Beijing Goodwill Metal Technology Co., Ltd. The sputtering gas was a mixture of Ar and O_2 (Ar:O_2 = 9:1 in pressure), and the RF power was 100 mW. (100)-oriented 20 × 20 × 0.5 mm^3 Si crystal was used as a substrate. The deposition rate of the films was 50–60 nm/h, and the thickness of the BTO thin film in this experiment was 327.8 nm, measured by the step profiler. Subsequently, the deposited film was annealed by conventional thermal annealing at 650 °C.

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A THz time-domain spectrometer (THz-TDS) system produced by Zomega Terahertz Corporation (USA) was used to measure the transmittance spectra of the film, as shown in Fig. 1. In the THz-TDS system, M1-M12 was reflective mirror and the delay line was composed of two reflective mirror. L1-L4 were polyethylene lenses. A fiber femtosecond laser beam was divided into two beams by a polarized beam splitter PBS. The two beams were named the pump beam and the probe beam, respectively. A half-wave plate (HWP) was placed to change the ratio between the pump beam and the probe beam. After passing through the attenuator and reflected by mirrors, the pump beam was focused on a GaAs photoconductive antenna for the generation of THz waves by lens L1. Meanwhile, the probe beam was focused into a GaAs crystal for the detection of the THz wave by lens L2. Confocal polyethylene lenses L3 and L4 were used to collimate and focus the emitted THz radiation onto the sample which was placed at the focus of L3 and L4. The transmitted THz wave was collected and focused on the GaAs crystal with two polyethylene lenses. The detectable frequency range was from 0.1 THz to 3 THz (3.3 cm^{-1} to 100 cm^{-1}). The frequency resolution was 4.5 GHz. In addition, an all-solid-state CW laser (center wavelength, 1064 nm) was employed for external optical pumping in this experiment. The light was obliquely incident upon the film at 60°, and the temperature of the system was maintained at 18 °C (291 K) during measurement.

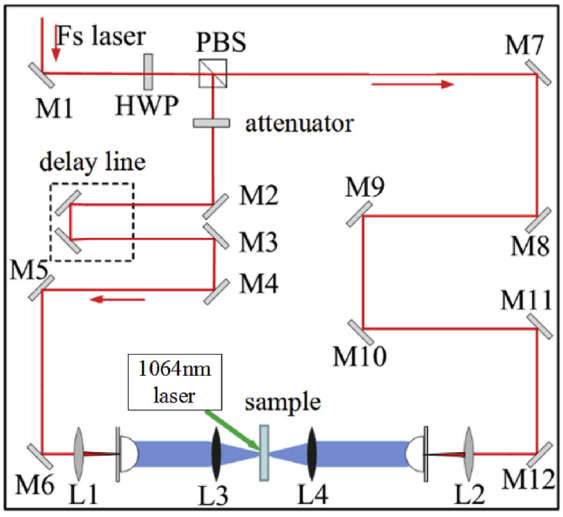

Fig. 1. Schematic diagram of the THz-TDS system. A 1064 nm laser was obliquely incident upon the surface of the film at 60° with regard to the polar axis.

3. Experimental results and discussion

Fig. 2 showed the time-domain signal waveforms of the sample obtained with different external optical pumping powers at room temperature. The time lag of the time-domain signal waveforms between the air and the substrate is 4.14 ps, as shown in Fig. 2(a). The signal waveform transmitted through sample shifted by only 0.12 ps compared with that of the bare substrate. In addition, compared with that without light excitation, the shift time of the signal waveform demonstrated minimal variations under increasing optical power. Furthermore, electric amplitude of the signal waveforms decreased to the minimum when the optical power reached to 400 mW.

Fig. 3 displayed the transmittance of the sample with BTO thin film, obtained from the ratio of the two waveforms via the equation: $t(\omega) = Es(\omega, P)/Er(\omega) = A\exp(-i\phi)$, where the $Es(\omega, P)$ was the frequency waveform of the bulk sample at a certain external optical pump power and $Er(\omega)$ was the frequency waveform of air. The transmittance of the sample was decreased with the increase of the external optical field, indicating that the transmittance was dependent on the optical pump power. According to the Fresnel format, the complex transmission function can be expressed as [18]:

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

Where t_f was the transmission of the BTO thin film and N_f, N_s are the complex refractive index of the film and the underlying substrate, respectively. d_f is the thickness of the ferroelectric film, and c is the speed of light. Hence, the complex dielectric constant $\varepsilon = N_f^2 = (n + i*k)^2$ could be retrieved from the THz transmission spectra.

Fig. 4 showed the complex dielectric constant of the BTO thin

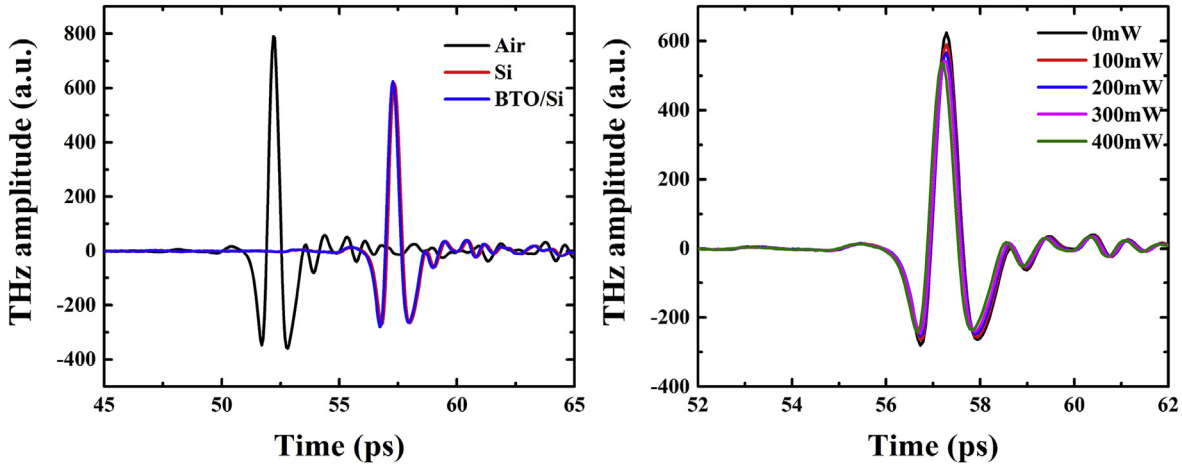

Fig. 2. (a) Time-domain signal waveforms of the whole sample, the substrate and air at room temperature. (b) was the time-domain signal waveforms of BTO thin film, respectively. The black line represented the data of the sample without optical pumping, the red line represented the data at P = 100 mW, the blue lines represent the data at P = 200 mW, the magenta line represented the data at P = 300 mW and the green line represented the data at P = 400 mW. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)