Influences of deposition parameters on microstructure and dielectric properties of BST thin film deposited by R.F. magnetron sputtering

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Abstract. Down graded barium strontium titanate (BST) thin films were prepared on Pt/Ti/SiO2/Si(100) substrate by radio frequency magnetron sputtering. The BST thin films were deposited at various substrate temperatures, then annealed at 700 °C for an hour. The influences of the deposition parameters on the crystallization behavior, microstructure and dielectric properties of BST thin films were investigated by X-ray diffraction, field emission scanning electron microscopy and dielectric frequency spectra. XRD results indicate that the BST thin films deposited at higher temperature have improved crystallization structure. The SEM observations show that the surface of the films is smooth with homogeneous grains. The dielectric properties of the films have been examined and discussed. The experiments show that BST thin films deposited at 650 °C, 3.0Pa working pressure exhibits superior dielectric properties: the highest dielectric constant is 448 and lowest dielectric loss is 0.013 at 100 kHz, respectively. These results make BST thin films be a promising candidate for microelectronic device application.

1. Introduction
Ba0.6Sr0.4TiO3 is thought to be the most promising candidate material of sensitive elements for uncooled infrared focal plane arrays (UFPA). The properties of Ba0.6Sr0.4TiO3 can be tailored for special applications including piezoelectric transducers and optical signal processing [1–4]. Recently, there have been some studies on the preparation of BST thin films. A variety of techniques has been used to prepare BST thin films [5]. Among these methods, radio frequency magnetron sputtering has obtained much interest because of its many advantages such as deposition from a single target, high deposition rate, low cost, large homogeneity and large area smooth film. However, very limited information of the influences of sputtering parameters on BST thin films has been reported, such as substrate temperature and sputtering pressure.

In this work, the influences of substrate temperature, sputtering pressure on the crystallization behavior, microstructure and dielectric property of compositionally graded BST thin films were studied.

2. Experimental
The n-type(100) silicon substrates were rinsed in acetone, ethanol, distilled water, and dried by nitrogen gas sequentially, then the substrate was thermally grown SiO2 about 100 nm, BST thin films with
different compositions were prepared on Pt/Ti/SiO_2/Si substrate by R.F.magnetron sputtering at substrate temperatures of 550°C, 600°C, 650°C. The sputtering was carried out at 0.5Pa, 3.0Pa, 6.5Pa, using a gas mixture of oxygen and argon. The compositionally graded BST thin films were achieved by continuously sputtering four different kinds of ceramic targets layer by layer with compositions of BaTiO_3, Ba_{0.9}Sr_{0.1}TiO_3, Ba_{0.8}Sr_{0.2}TiO_3 and Ba_{0.7}Sr_{0.3}TiO_3, respectively. All the films were annealed at 700 °C for an hour, then they were cooled down to room temperature naturally. X-ray diffraction (XRD) technique was carried out to identify the crystal structure of the compositionally graded BST thin films by using a D/max-ⅢC diffractometer (Cu Kα radiation with a working voltage of 35 kV and current of 25 mA, wave-length $\lambda = 0.154176$ nm, scan speed of 10°/min, and scan range 20–67°). The microstructure of the graded BST thin films was examined by scanning electron microscopy (JOEL JSM 6700F). To measure the dielectric property, Pt top electrode with diameter of 300 μm was deposited on the top surface of the graded BST thin films by DC sputtering. The dielectric constant and dielectric loss were measured by an Agilent 4294A impedance analyzer.

3. Results and discussion

3.1. The effect of deposition parameters on the crystallization of BST thin films

Fig.1 was XRD patterns of graded BST thin films deposited at different temperature and sputtering pressure. It can be seen that the intensity of (100), (110), and (111) peaks of the BST thin films enhance with the increase of the deposition temperature. (100) and (200) peaks become sharper and more intense at 600°C, which reveal that the better crystallinity of the BST thin films can be obtained at higher substrate temperature.

The temperature dependence of the film quality can be interpreted mainly by the mobility of the particles at different temperature, the particles with low surface mobility will prevent the crystallization of the thin films at low temperature [6]. The dominant diffraction peaks of BST thin films deposited at 3.0Pa are (110) and (111). (110) peak intensity decreases apparently when the sputtering pressure increases up to 6.5 Pa, which indicates that the crystallizability of the BST thin films is not very well at 6.5pa.

![Fig 1. XRD patterns of graded thin films deposited at different temperature(a) and sputtering pressure(b)](image)

3.2. The effect of deposition parameters on Microstructure of BST thin films

The microstructures of the graded BST thin films as a function of substrate temperature and sputtering pressure were examined by SEM images. It can be observed from Fig.2 that the surface morphologies vary with the change of the substrate temperature and the sputtering pressure. While deposited at 600°C, sputtering pressure was 3.0pa, the films have smooth and densely packed structure with homogeneous grains, the grain size is about 70nm, which is showed in Fig.2.(b),(e). This is due to that the mobility of species becomes higher and induces a higher speed of the coalescence of grain islands as the substrate temperature increases. Further increasing the pressure to 6.5pa, the graded BST grains do not grow remarkably and the roughness of graded BST thin film increases to 6.32nm.
3.3. The effect of deposition parameters on the dielectric properties of BST thin films

The dielectric properties of the graded BST thin films were measured by a metal-ferroelectric-metal (MFM) configuration, in which the graded films were sandwiched between bottom and top Pt electrodes. The frequency dependence of dielectric constant of graded BST thin films deposited at different substrate temperature and different sputtering pressure is shown in Fig.3(a),(b). It’s found that the dielectric constant of all the samples decrease with the increase of the frequency. The decrease of the dielectric constant may be caused by the accumulation of space charge that involves lots of oxygen vacancies in the graded BST thin films. The experiments show that the highest dielectric constant and the lowest dielectric loss at 100KHz of the graded BST thin films deposited at 650°C and 3.0 Pa are 448,0.013, respectively, which exhibit excellent dielectric property of the graded BST thin films.

**Fig 2.** SEM images of compositionally graded BST thin films deposited at different substrate temperatures: (a) 550 °c; (b) 600 °c; (c) 650 °c; (d) 0.5pa; (e) 3.0pa; (f) 6.5pa

**Fig 3.** Dielectric properties of compositionally graded BST thin films as function of frequency, (a) deposited at different temperature; (b) deposited at different sputtering pressure.
4. Conclusions
Compositionally graded BST thin films with good crystallization structure were successfully fabricated by RF magnetron sputtering technique at a higher temperature of 600°C and in the sputtering pressure of 3.0Pa, the dominant X-ray diffraction peaks (100),(200) became sharper and more intense. The graded BST thin films deposited at 650°C and 3.0Pa possess the highest dielectric constant 448 and the lowest dielectric loss 0.013 at 100kHz, which exhibit excellent dielectric property of the BST thin films. These results showed that the deposition parameters have important effect on the crystallization, microstructure and dielectric properties of the BST thin films.

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