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Effect of bottom electrodes on the dielectric properties of barium strontium titanate thin films

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Abstract

Barium strontium titanate ($Ba_{0.5}Sr_{0.5}TiO_3$, BST) thin films were prepared on Pt/Ti/SiO₂/Si and Pt/Cr/SiO₂/Si substrates by sol-gel method. The effects of Pt/Ti and Pt/Cr bottom electrodes on the microstructure and dielectric properties of BST thin films were studied. The X-ray diffraction patterns show that both these films crystallize into a perovskite structure. The atomic force micrographs indicate that the films using Pt/Ti and Pt/Cr as bottom electrodes have different grain distributions and grain sizes. The dielectric constant of the films using Pt/Cr as bottom electrodes decreases significantly and the loss tangent and tunability decrease slightly. However, the figure of merit of the films using Pt/Cr as bottom electrodes still increases from 24.3 to 27.6.

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Keywords: Sol-gel method; Barium strontium titanate; Bottom electrodes; Dielectric properties

1. Introduction

Barium strontium titanate ($Ba_{1-x}Sr_{1-x}TiO_3$, BST) thin films have been widely investigated for tunable microwave devices such as tunable filters, tunable oscillators, phase shifters and phased array antennas due to their high tunability and low loss tangent [1-3]. It is well known that the dielectric properties of BST thin films are greatly dependent on preparation methods and film microstructure. However, the dielectric properties are also dependent on bottom electrodes. Generally, platinum is used as a standard electrode for integration of ferroelectric thin films with silicon because of its low resistivity, high work function and good stability in the perovskite crystallization temperature. To ensure a good Pt adhesion onto the silicon layer, a titanium thin layer is generally used. It has been reported that during deposition of ferroelectric thin films at high temperature, Ti can migrate into the surface via Pt grain boundaries and may give rise to the formation of hillocks [4-7]. The formation of hillocks can greatly undermine the

dielectric properties and hinder practical applications of ferroelectric thin films.

However, few published work on BST thin films use Pt/Cr as bottom electrodes. In this study, $Ba_{0.5}Sr_{0.5}TiO_3$ thin films were prepared on Pt/Ti/SiO₂/Si and Pt/Cr/SiO₂/Si substrates by sol-gel method. The effects of Pt/Ti and Pt/Cr bottom electrodes on the microstructure and dielectric properties were examined.

2. Experimental details

The starting materials were barium acetate, strontium acetate and titanium-tetrabutoxide. Glacial acetic acid and 2-methoxyethanol were used as solvents. Barium acetate and strontium acetate powders in the mole ratio of 1:1 were dissolved in heated glacial acetic acid (HAc) and the mole ratio of (Ba+Sr) to HAc is 1:12. Then, a stoichiometric amount of titanium-tetrabutoxide was diluted in 2-methoxyethanol and added to the solution of barium acetate and strontium acetate while stirring. Ethylene glycol and deionized water were added to the above solution and stirred to get a clear precursor solution. The mole ratio of ethylene

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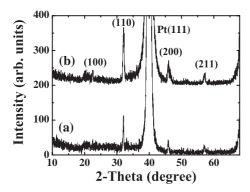
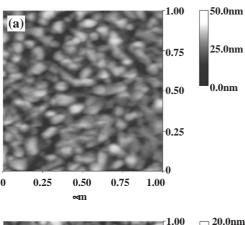


Fig. 1. XRD patterns of BST thin films on (a) Pt/Ti/SiO $_2$ /Si and (b) Pt/Cr/SiO $_2$ /Si substrates.

glycol to HAc is 1:3 and the mole ratio of de-ionized water to titanium-tetrabutoxide is 3:2. The final concentration was adjusted to 0.45 M by adding 2-methoxyethanol. The precursor solution was spun on Pt/Ti/SiO₂/Si and Pt/Cr/SiO₂/Si substrates at 2500 rpm for 30 s. Each layer of the wet film was dried at 80 °C for 30 min and subsequently pyrolysed at 400 °C for 30 min. This step was repeated several times to obtain the desired thickness. The film was finally annealed at 750 °C for 1.5 h.

The structure of the films was analyzed by X-ray diffraction technique (XRD, Rigaku D/max) using CuKα



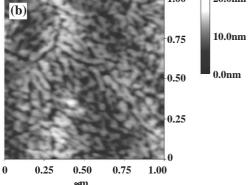


Fig. 2. AFM surface micrograph of BST thin films on (a) Pt/Ti/SiO $_2$ /Si and (b) Pt/Cr/SiO $_2$ /Si substrates.

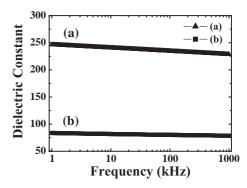


Fig. 3. The frequency dispersion of the dielectric constant of BST thin films on (a) Pt/Ti/SiO₂/Si and (b) Pt/Cr/SiO₂/Si substrates.

radiation. The surface micrograph was investigated by an atomic force micrograph (AFM, Nanoscope IIIa, Digital Instruments) using contact mode with a standard silicon nitride tip. To study the dielectric properties, 0.4-mm-diameter Pt circular electrodes were sputtered onto the films to form a metal-ferroelectric-metal configuration. The dielectric properties were measured using a Precision Impedance Analyzer (Agilent 4294A).

3. Results and discussion

Fig. 1 shows the XRD patterns of BST thin films on Pt/Ti/SiO₂/Si and Pt/Cr/SiO₂/Si substrates. It is evident that both these films are perovskite polycrystalline without preferential orientation. It is also found that the full width at half-maximum of the dominant (110) peak of the films using Pt/Ti as bottom electrodes is smaller than that of the films using Pt/Cr as bottom electrodes. The results indicate that the films using Pt/Ti as bottom electrodes have a larger grain size than the films using Pt/Cr as bottom electrodes. It has been reported that when a Pt/Ti bilayer is used as a bottom electrode for lead zirconate titanate (PZT) thin films, Ti atoms diffuse out through the grain boundaries of the Pt layer onto the Pt surface, and the out-diffused Ti facilitates the

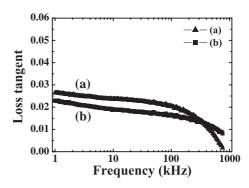


Fig. 4. The frequency dispersion of the loss tangent of BST thin films on (a) $Pt/Ti/SiO_2/Si$ and (b) $Pt/Cr/SiO_2/Si$ substrates.

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