Available online at www.sciencedirect.com

SCIENCE DIRECT

Thin Solid Films 485 (2005) 82 – 89

www.elsevier.com/locate/tsf

# Effects of oxygen pressure on lattice parameter, orientation, surface morphology and deposition rate of $(Ba_{0.02}Sr_{0.98})TiO_3$ thin films grown on MgO substrate by pulsed laser deposition

Can Wang[1], B.L. Cheng*, S.Y. Wang, H.B. Lu, Y.L. Zhou, Z.H. Chen, G.Z. Yang

*Institute of Physics, Chinese Academy of Sciences, Beijing 100080, PR China*

Received 15 August 2004; received in revised form 25 January 2005; accepted 24 March 2005
Available online 23 May 2005

**Abstract**

$(Ba_{0.02}Sr_{0.98})TiO_3$ thin films were grown on MgO substrate by pulsed laser deposition (PLD) techniques at various oxygen pressures from 40 to $10^{-3}$ Pa. Effects of oxygen pressure on lattice parameter, orientation, surface morphology and deposition rate of the thin films were investigated. X-ray diffraction patterns indicate that, with decreasing oxygen pressure, crystal orientation of the thin films develops from (110) preferred orientation to random orientation, and finally to perfectly (00l) texture. Meantime, lattice parameter of the thin films increases with decreasing oxygen pressure. Especially, perfectly (00l)-oriented $(Ba_{0.02}Sr_{0.98})TiO_3$ thin film was obtained under oxygen pressure of $10^{-3}$ Pa at substrate temperature of 850 °C, in spite of the large lattice mismatch of about 8% between $(Ba_{0.02}Sr_{0.98})TiO_3$ and MgO. Atomic force microscopy images show that all the thin films have granular structure and the thin film grown at $10^{-3}$ Pa has much more homogeneous grain size distribution compared to all the other thin films. Moreover, the films grown at oxygen pressures of 1 and $10^{-1}$ Pa have more smooth surfaces, and the deposition rate of thin film increases with decreasing oxygen pressure.


*PACS:* 77.55; 68.55
*Keywords:* Oxygen pressure; Pulsed laser deposition; BST thin film; Structural characterization

## 1. Introduction

Thin films of $Ba_xSr_{1-x}TiO_3$ (BST) have recently been considered to be important materials for tunable microwave devices such as microwave tunable phase shifters, tunable filters, and high-$Q$ resonators for radar and communication applications [1–3]. Tunable ferroelectric devices are generally used above the Curie temperature in paraelectric regime to reduce voltage hysteresis inherent to ferroelectric phase, and to lower dielectric loss because the ferroelectric phase introduces substantial intrinsic dielectric loss. BST thin films are considered to be the best candidate for this application because of their large electric field tunability, relatively low dielectric loss and variable Curie temperature. Transition temperature of BST from paraelectric to ferroelectric phase can be changed from 400 to 30 K by varying ratio of Ba/Sr, and it is favorable to obtain BST in paraelectric phase with good properties for use in specific temperature [4,5]. For example, to integrate high temperature superconductor thin films for high frequency system, high Sr composition BST should be selected for the microwave devices used at low temperature [1].

For the tunable microwave devices, it is very important to grow a thin film with high tunability and low dielectric loss. MgO is one of the best substrates for microwave applications because of its excellent dielectric properties such as a lower dielectric loss of about $3\times10^{-7}$ and dielectric constant of about 9.7. These physical parameters of MgO compare well with those of $Al_2O_3$ ($\varepsilon\approx10$, tan $\delta\approx6\times10^{-5}$) or $LaAlO_3$ ($\varepsilon\approx25$, tan $\delta\approx6\times10^{-5}$) for microwave device applications. Unfortunately, there is a

* Corresponding author. Fax: +86 10 82649531.
*E-mail address:* blcheng@aphy.iphy.ac.cn (B.L. Cheng).
[1] Present address: Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge, CB2 3EQ, UK.

0040-6090/$ - see front matter © 2005 Published by Elsevier B.V.
doi:10.1016/j.tsf.2005.03.055

large lattice misfit between BST (0.3905 nm for $SrTiO_3$) and MgO (0.4216 nm) substrate, especially in BST with rich Sr composition because the lattice constant of BST decreases with increasing Sr content, and also there is a difference in the thermal expansion coefficient ($14\times10^{-6}$/K for MgO and $11\times10^{-6}$/K for $SrTiO_3$). Therefore, fabrication of high quality BST films on MgO substrate became a challenge. Pulsed laser deposition (PLD) technique has been regarded as one of the best methods for numerous advantages, including film stoichiometry close to the target, reproducibility, uniformity and simplicity, so it has been widely used in preparing ferroelectric thin films. Chen et al. [6] have reported that epitaxial $Ba_{0.6}Sr_{0.4}TiO_3$ thin films on MgO can only be achieved at temperature higher than 820 °C and under oxygen pressure in a narrow range between 200 and 300 mTorr. Kalyanaraman et al. [7] have investigated the growth of $SrTiO_3$ thin films on MgO substrates, and their results show that high quality thin films can only be grown at proper optimization temperature and oxygen pressure. Delage et al. [8] also investigated the microstructure and dielectric properties of BST thin films grown on MgO by PLD with and without buffer layer. Though many studies have reported BST thin film grown by PLD technique, systematic investigation on Sr-rich BST thin films on MgO substrate is still necessary to understand growth mechanism for fabricating high quality thin films.

As we know, oxygen vacancies, which affect the microstructure and properties of BST thin films, are introduced inevitably in the PLD-deposited oxide thin films for the lower deposition oxygen pressure. The lower the oxygen pressure, the higher the content of oxygen vacancies [9]. Moreover, existence of oxygen gas in the ambient has a strong influence on expansion of the laser-produced plasma plume and modifies the kinetic energy of the ablated species, resulting in difference on characteristics of thin film, such as surface morphology, growth rate, orientation, stoichiometric composition, etc. [10–14]. In this paper, elaboration and characteristics of $Ba_{0.02}Sr_{0.98}TiO_3$ films on MgO by PLD under a larger range of oxygen pressures are reported. It is found that lattice parameters, orientation, surface morphology and deposition rate of the thin films are sensitive to the ambient oxygen pressure.

## 2. Experimental details

Thin films of $(Ba_{0.02}Sr_{0.98})TiO_3$ were prepared by PLD technique employing a XeCl excimer laser with wavelength 308 nm, pulsed with 20 ns and frequency of 4 Hz. The laser with energy density of about 2 J/cm$^2$ was focused on ceramic target mounted on a motor-driving rotary stand rotating at a constant speed to ensure a uniform ablation rate. The ceramic target was prepared through conventional solid-state reaction process, using $BaCO_3$, $SrCO_3$ and $TiO_2$ with an analytic reagent grade of purity. Raw materials with stoichiometric composition of $(Ba_{0.02}Sr_{0.98})TiO_3$ were milled and then calcined at temperature of 950 °C. The calcined powders were pulverized and pressed into disk, then sintered at temperature of 1420 °C for 20 h. The thin films were grown on MgO (001) substrates with a distance of 4.5 cm between the substrate and target at 850 °C under oxygen pressures of 40, 10, 1, $10^{-1}$, $10^{-2}$ and $10^{-3}$ Pa, respectively.

Crystallographic characteristics of the BST thin films were analyzed by X-ray diffraction (XRD) employing Cu Kα radiation (40 kV, 50 mA). Atomic force microscopy (AFM) was used to measure the surface morphology and roughness of the films, a randomly chosen area of $1\times1$ μm$^2$ on each film was surveyed. Thickness of the BST thin films was measured by a surface profile measuring system (DEKTAK III, USA) at a step grown by putting a mask on part of the substrate during deposition process.

## 3. Results and discussion

### 3.1. XRD patterns

Fig. 1 shows the XRD patterns of $(Ba_{0.02}Sr_{0.98})TiO_3$ thin films on MgO substrates deposited by PLD at oxygen pressures of 40, 10, 1, $10^{-1}$, $10^{-2}$ and $10^{-3}$ Pa, respectively. As revealed from the XRD patterns, all peaks indexed in Fig. 1 correspond to perovskite phase, which indicates all $(Ba_{0.02}Sr_{0.98})TiO_3$ thin films well crystallized into single phase. Moreover, with decreasing growth oxygen pressure, intensities of (001) and (002) diffraction peaks are

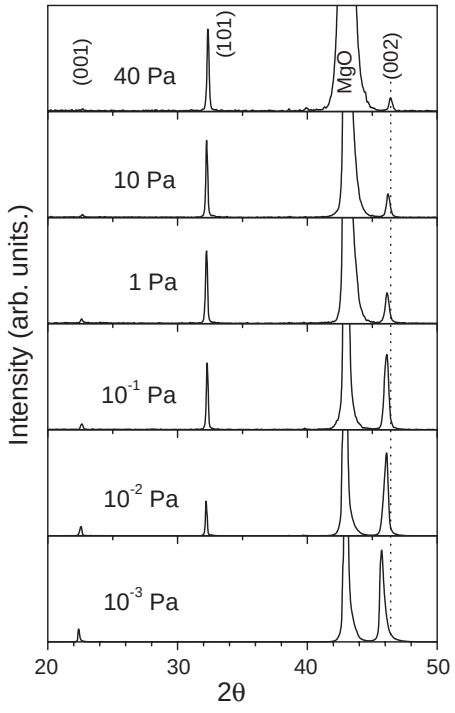


Fig. 1. XRD patterns of $(Ba_{0.02}Sr_{0.98})TiO_3$ thin films grown on MgO at various oxygen pressures by PLD.