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Thin Solid Films 510 (2006) 115–118
thin solid films
www.elsevier.com/locate/tsf

Microwave dielectric properties of W-doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films grown on (001) MgO by pulsed laser deposition with a variable oxygen deposition pressure

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Received 12 December 2005; accepted 16 December 2005
Available online 19 January 2006

Abstract

The microwave dielectric properties of $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped thin films deposited using pulsed laser deposition, are improved by a novel oxygen deposition profile. The thin films were deposited onto (001) MgO substrates at a temperature of 720 °C. A comparison is made between three different oxygen ambient growth conditions. These include growth at a single oxygen pressure (6.7 Pa) and growth at two oxygen pressures, one low (6.7 Pa) and one high (46.7 Pa). Films were deposited in a sequence that includes both a low to high and a high to low transition in the oxygen deposition pressure. Following deposition, all films were post-annealed in 1 atm of oxygen at 1000 °C for 6 h. The dielectric Q (defined as $1/\tan\delta$) and the dielectric constant, ε_r, were measured at room temperature, at 2 GHz, using gap capacitors fabricated on top of the dielectric films. The percent dielectric tuning (defined as $(\varepsilon_r(0\text{ V})-\varepsilon_r(40\text{ V}))/\varepsilon_r(0\text{ V})\times 100$) and figure of merit (FOM) (defined as percent dielectric tuning $\times Q(0\text{ V})$) were calculated. The film deposited using the two-stage growth conditions, 6.7/46.7 Pa oxygen, showed a maximum $Q(0\text{ V})$ value with high percent dielectric tuning and gave rise to a microwave FOM twice as large as the single stage growth condition. The improved dielectric properties are due to initial formation of a film with reduced interfacial strain, due to the formation of defects at the film/ substrate interface resulting in a high $Q(0\text{ V})$ value, followed by the reduction of oxygen vacancies which increases the dielectric constant and tuning.

PACS: 81.15.Fg; 84.40.Lj; 77.84.Dy; 68.55
Keywords: Pulsed Laser Deposition (PLD); Microwave properties; BST; Dielectric properties

1. Introduction

$Ba_xSr_{(1-x)}TiO_3$ $(0 \le x \le 1)$ (BST) thin films have been extensively investigated as candidates for development of a new class of tunable microwave signal processing devices [1–3]. The films exhibit an electric-field dependent dielectric constant and have been used to fabricate tunable capacitors for use in microwave varactors and phase shifters. However, the relatively high loss tangent ($\tan\delta$) of BST films limits their use. Oftentimes it is observed that some materials exhibit a large electric-field induced change in the dielectric constant, but also a high dielectric loss, while other materials exhibit a very small tuning, and at the same time a low dielectric loss. The current challenge is to optimize the processing of the material such that it exhibits a large electric-field induced tuning, with a minimum dielectric loss.

Two of the major factors affecting the dielectric loss of BST thin films are: a) strain in the film caused by the film-substrate thermal and lattice mismatch [4]. Those can be changed by varying the oxygen pressure during deposition [5–8] and/or

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doi:10.1016/j.tsf.2005.12.270

post-anneal treatment [9,10] and b) the presence of free carriers in the BST film due to the presence of oxygen vacancies, which increases microwave losses. Several dopants have been used to minimize free carriers and reduce microwave losses in BST thin films [11–14]. W acts as a donor dopant in BST and was found to reduce loss tangent values [15–17]. $Ba_{0.6}Sr_{0.4}TiO_3$ containing 1 mol% W films deposited at 6.7 Pa (50 mTorr) oxygen pressure on (001) MgO substrates have shown high dielectric figure of merit (FOM) [8], defined as $(\varepsilon_r(0\ V) - \varepsilon_r(40\ V))/\varepsilon_r(0\ V) \times 100 \times Q(0\ V)$, where $\varepsilon_r(0\ V)$ is the film dielectric constant at 0 V, $\varepsilon_r(40\ V)$ is the film dielectric constant at 40 V, $Q(0\ V)$ is the dielectric quality factor and is defined as $1/\tan\delta$ at 0 V.

A post-deposition anneal of BST films deposited on (001) MgO substrates, with oxygen ambient pressure, was found to increase the dielectric Q, by reducing the strain between the film and the substrate. However, it is often observed that while increasing the dielectric Q, the post-deposition anneal also leads to a decrease in the electric field induced tuning [9]. The inverse relationship between the loss tangent and dielectric tuning in strained BST films can be minimized by depositing a buffer layer between the crystalline BST film and the crystalline (001) MgO substrate [4,17–19].

The objectives of this work is to introduce a unique, continuous, two oxygen deposition pressure stages procedure, for increasing the electric-field induced change in the dielectric properties of pulsed laser deposition [20,21] grown $Ba_{0.6}Sr_{0.4}TiO_3$ thin films containing 1 mol% W films, while at the same time preserving a high dielectric Q. A tentative mechanism for strain relief and oxygen vacancies reduction is suggested.

2. Experimental

A pulsed, KrF excimer laser (248 nm, 30 ns Full Width at Half Maximum (FWHM)) operating at 10 Hz was used to deposit 0.8–1.2 μm thick, $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped films over (001) oriented MgO single crystals, as described previously [8]. The laser beam was focused with a 50 cm focal length lens onto a rotating $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped target purchased from Target Materials Inc. For all depositions the (001) MgO substrate temperature was maintained at 720 °C. Single-stage films were deposited at 6.7 Pa oxygen pressure with about 10,000 laser pulses since it was previously reported [8] that $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped films deposited at 6.7 Pa oxygen pressure on (001) MgO substrate showed maximum dielectric FOM compared to films deposited at other oxygen deposition pressures. Hence no film was made using single-stage oxygen pressures other than 6.7 Pa for the comparison to two-stage oxygen pressures process.

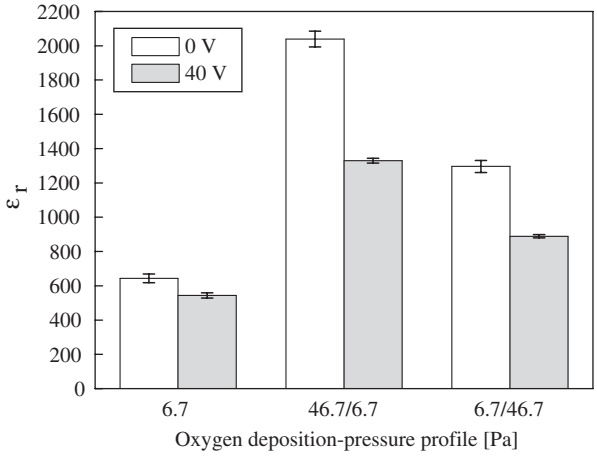

Fig. 1. Average dielectric constant at 2 GHz for $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped films deposited in different oxygen-pressure profiles.

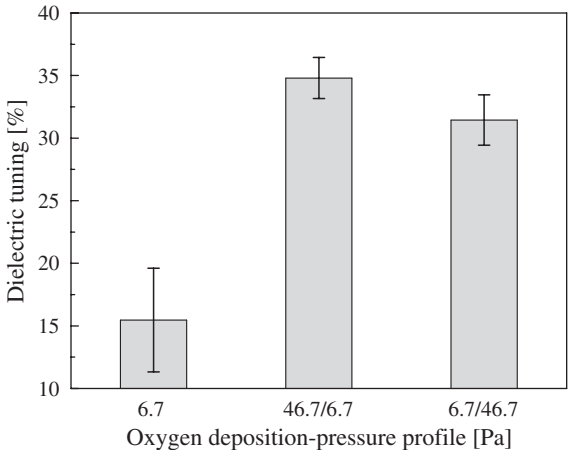

Fig. 2. Average percent dielectric tuning at 2 GHz for $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped films deposited in different oxygen-pressure profiles.

The two-stage oxygen deposition pressures were either a) 46.7 Pa for 12,000 laser pulses, then the oxygen deposition pressure reduced to 6.7 Pa for the next 13,000 laser pulses or b) 6.7 Pa for 12,500 laser pulses, then the oxygen deposition pressure increased to 46.7 Pa for the next 12,5000 laser pulses. The laser was operating while the oxygen pressures were changed. The film thickness was measured using a Tencor P-10 surface profilometer. All films were post-annealed in oxygen ambient pressure at 1000 °C for 6 h.

After the post-deposition anneal, an eight-gap capacitor pattern (5 μm gap width), in various orientations, were fabricated on top of the films by standard photolithography. A thin Cr adhesion layer (~100 Å) was used for the gold capped silver electrode. The silver was deposited in an e-beam evaporator over the patterned resist. Lift-off was then used to delineate the gap capacitor pattern. Thick silver (2–5 μm) was used to reduce the conductor losses at microwave frequencies (1–20 GHz). The dielectric properties of the deposited films were measured at 2 GHz at room temperature using an HP 8510 network analyzer and cascade probes. The capacitance and Q of the gap caps were calculated from the measured S11 parameters of the caps using a parallel resistor–capacitor circuit model. The conformal mapping technique [22] for the analysis of gap capacitors on layered substrates was used to extract the dielectric constants of the $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped films from the calculated capacitance values.

The crystal structure (ω scan) of as-deposited $Ba_{0.6}Sr_{0.4}TiO_3$ 1 mol% W-doped films on (001) MgO, at 720 °C, single-stage