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Processing and properties of BST thin films for tunable microwave devices

Jin Xu*, Wolfgang Menesklou, Ellen Ivers-Tiffée

Institut für Werkstoffe der Electrotechnik, Universität Karisruhe (TH), 76131 Karlsruhe, Germany

Abstract

Dielectric nonlinearity and low losses of ferroelectric thin films with perovskite structure have made them prospective candidates for electric field tunable microwave devices. In this paper, we present the correlation between preparation conditions, microstructure and dielectric behavior of $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) thin films deposited by RF magnetron sputtering on (001) MgO and $LaAlO_3$ single crystal substrates. The composition, crystallinity and morphology of the thin films were analysed by Rutherford backscattering (RBS), X-ray diffraction (XRD) and transmission electron microscopy (TEM), respectively. The dielectric measurements were carried out using interdigital capacitors (IDC) with Au electrodes on thin films at 1 MHz. By optimizing the processing, a tunability $\{\text{defined as tu\%} = [\varepsilon(0) - \varepsilon(E_{max})]/\varepsilon(0)\}$ of 22% at $E_{max} = 10$ kV/mm and a low loss tangent of 0.0023 have been achieved. In addition, the role of mismatch stress (both compressive and tensile) on the dielectric properties of the thin films is also discussed.

Keywords: $BaTiO_3$ and titanates; Dielectric properties; RF magnetron sputtering; Thin films

1. Introduction

The high dielectric nonlinearity (manifested as the strong dependence of dielectric constant on electric field) of ferroelectric materials with perovskite structure have made them prospective candidates for microwave applications.[1,2] In recent years, a considerable amount of work has been done in the area of developing tunable ferroelectric thin films for fabricating electric field tunable microwave devices, such as tunable oscillators, tunable filters, phase shifters and varactors,[3–5] which can be used in radar and communication systems operating at room temperature. The realization of high performance tunable microwave devices based on ferroelectric thin films with combination of low losses and high dielectric tunability $(tu\% = (\varepsilon(0) - \varepsilon(E_{max}))/\varepsilon(0))$ will have a significant impact on wireless communications, including satellite applications.

Nowadays, the common studies in this field are mainly focused on $Ba_xSri_{1-x}TiO_3$ (BST) system.[3–7] Regardless of deposition method and substrate type, the critical properties of the thin films which need to be optimized are the magnitude of tunability as a function of applied electric field and dielectric losses.

In this paper, the investigation of $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) thin films prepared using various sputtering conditions is described. The correlation between mismatch strain and thin film dielectric properties has been investigated, and a conclusion of BST films deposited on MgO substrates under tensile stress show higher tunability values than those deposited on $LaAlO_3$ substrates under compressive stress can be given. The effect of post-annealing on the dielectric properties of BST thin films measured at 1 MHz was also presented.

2. Experimental

BST thin films with the thickness of 350– 500 nm were prepared by RF magnetron sputtering on (001) MgO and $LaAlO_3$ single crystal substrates. A 100 mm diameter target (a sintered stoichiometric $Ba_{0.5}Sr_{0.5}Ti0_3$ ceramic disk) was positioned 35 mm oppositely away from the substrate ("on-axis" sputtering). Thin films were deposited using an Ar–O_2 gas mixture of 2.5–8.5×10^{-2} mbar and at a substrate temperature (T_s) of

* Corresponding author. Tel.: +49-721-608-8148; fax: +49-721-608-7492.

E-mail address: jin.xu@iwe.uni-karlsruhe.de (J. Xu).

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600–700 °C. The sputtering condition is shown in Table 1. In order to optimizing the preparation processing, post-annealing in air at 900 °C for 5 h was also performed for some BST films.

The thickness, composition, crystallographic orientation, microstructure and surface roughness of BST thin films were characterized using Rutherford backscattering (RBS), X-ray diffraction (XRD) transmission electron microscopy (TEM), and atomic force microscopy (AFM), respectively.

Dielectric properties (dielectric constant ε_r, loss tangent $\tan\delta$ and tunability tu%) of BST thin films as a function of applied electric field and temperature were measured at 1 MHz. To obtain the values of ε_r and $\tan\delta$, the capacitance and losses measurements of interdigital capacitors (IDC) were carried out, which was described elsewhere.[8] The IDC, as shown in Fig. 1, were fabricated on BST thin film by evaporating a 10 nm chromium adhesion film and a 100 nm thick gold metallization, followed by a standard lift-off process. A modified conformal-mapping technique and a partial capacitance method were applied to evaluate the dielectric properties.[9]

3. Result and discussion

3.1. Crystal structure, composition and microstructure of BST thin films

The relationship between the various deposition conditions and the crystal structure of the BST thin films were studied. The influence of substrate temperature on crystallographic orientation, and the significant effect of sputtering gas ratio on out-of-plan orientation are shown in Figs. 2 and 3, respectively.

Table 1
Sputtering conditions of BST thin films

Target-substrate distance	35 mm
Substrate temperature	600–700 °C
Sputtering gas	O_2: Ar = 10 sccm :90 sccm
Total pressure of sputtering gas	$2.5-8.5\times10^{-2}$ mbar
Plasma power	RF 100 W or 150W
Growth rate	3.83 nm/min
Composition determined by RBS	$Ba_{0.5}Sr_{0.5}Ti_{0.96}O_3$

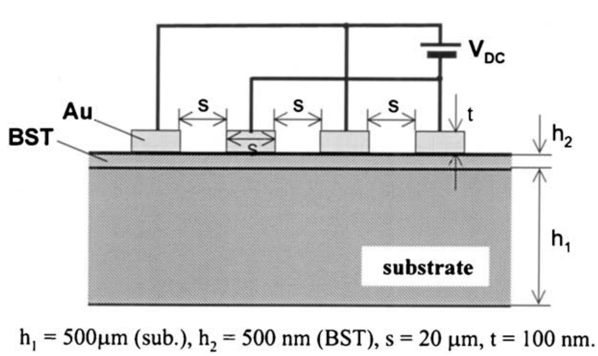

Fig. 1. Cross-section structure of IDC.

It is well known that the increasing of the substrate temperature can improve the epitaxial orientation of the thin films. From the XRD pattern shown in Fig. 2, we found that the BST thin film deposited at a T_s as low as 600 °C has a poor quality of crystallization, while film deposited at 700 °C exhibits a good c-axis orientation. For BST films deposited on $LaA1O_3$ at 600, 650, and 700 °C, the full width at half-maximum (FWHM) of the ω-scan peaks for BST (200) reflection were determined as 1.252, 0.285 and 0.136°, respectively, which indicated a better out-of-plane orientation appearing with the increasing of T_S.

The effect of sputtering gas ratio on out-of-plan orientation is clearly shown in Fig. 3. It can be seen that the appropriate ratio of Ar:O_2 should be 90:10–90:20. The corresponding $\theta-2\theta$ scan patterns also show better crystallization of the BST films prepared in this gas ratio range.

BST thin films grown under optimized sputtering conditions were characterized by XRD to be single phase and exclusively *c*-axis epitaxial. The typical film compositions determined by RBS (~5% deviation of evaluation) were relatively constant, as $Ba_{0.5}Sr_{0.5}Ti_yO_3$ ($y = 0.96–1.03$), indicated the proper composition of the films.

Fig. 4 shows a cross-sectional TEM image and the selected-area-diffraction (SAD) pattern of the BST thin film grown on MgO substrate. No columnar grain morphology and visible grain boundary can be observed in TEM, and SAD pattern shows relatively integrated cubic crystal structure.

A root-mean-square (rms) roughness of 3 nm was determined from the AFM image for the typical BST film, which demonstrates the smoothness of the film surface.

3.2. Dielectric properties

The dielectric constant (ε_r) and loss tangent ($\tan\delta$) as a function of the temperature for optimized BST thin film are plotted in Fig. 5. It is obvious that the dielectric constant (ε_r) is rather low temperature dependent (which is significantly different from the corresponding bulk materials) but high electric field dependent. At room temperature, a moderate dielectric constant value of 294 and a low loss tangent of 0.0024 were obtained at $E = 0$ kV/mm, and a tunability as high as 22% can be achieved at $E_{max} = 10$ kV/mm.

The dielectric properties of various BST thin films are listed in Table 2. Fig. 6 shows the influence of substrates and post-annealing on electric field dependent tunability.

It can be noticed that the BST thin films deposited at low temperature ($T_s = 600$ °C) exhibit a high loss tangent and a very low tunability, which can be explained by the poor crystal quality indicated in 3.1. In general, the as-deposited BST thin films grown on MgO shows a lower dielectric constant and a higher tunability than those on $LaAlO_3$. After post-annealing in air at 900 °C for 5 h, the dielectric constant increase slightly, and the tunability increase significantly for BST thin films grown on both substrates.

The proposed reason for the different dielectric behavior of the BST thin films deposited on MgO and $LaAlO_3$ substrates may be the different film stress, which arises from the lattice mismatch and the different thermal expansion coefficients between the thin film and the substrate. Some related parameters are summarized in Table 3.

It is apparent that the BST thin films deposited on MgO substrates are under tensile stress ($\alpha_{MgO} = 0.421$ nm $> \alpha_{BST} = 0.395$ nm), and that those deposited on $LaAlO_3$ substrates are under compressive stress ($\alpha_{LaAlO3} = 0.379$ nm $< \alpha_{BST} = 0.395$ nm) (*α: lattice constant). The stress in the thin film induces an important effect to the dielectric behavior.

It was known that the as-deposited BST films contained significant oxygen vacancies.[10] The process of post-annealing in air or O_2 is reducing these oxygen vacancies. Therefore, the dielectric properties can be obviously improved, and according to this approach, a higher tunability can be achieved.

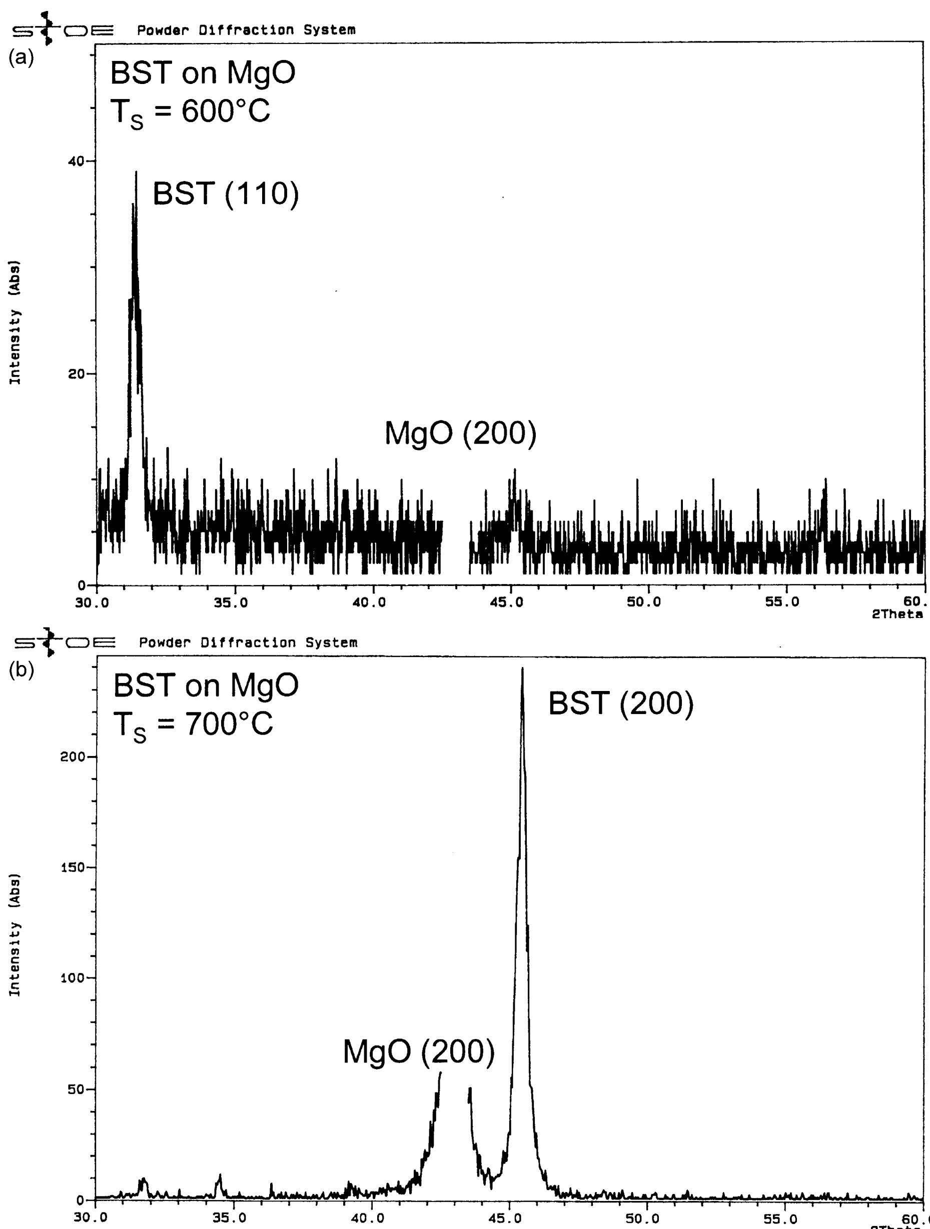

Fig. 2. XRD analysis of BST thin films deposited at T_S of (a) 600 °C and (b) 700 °C.

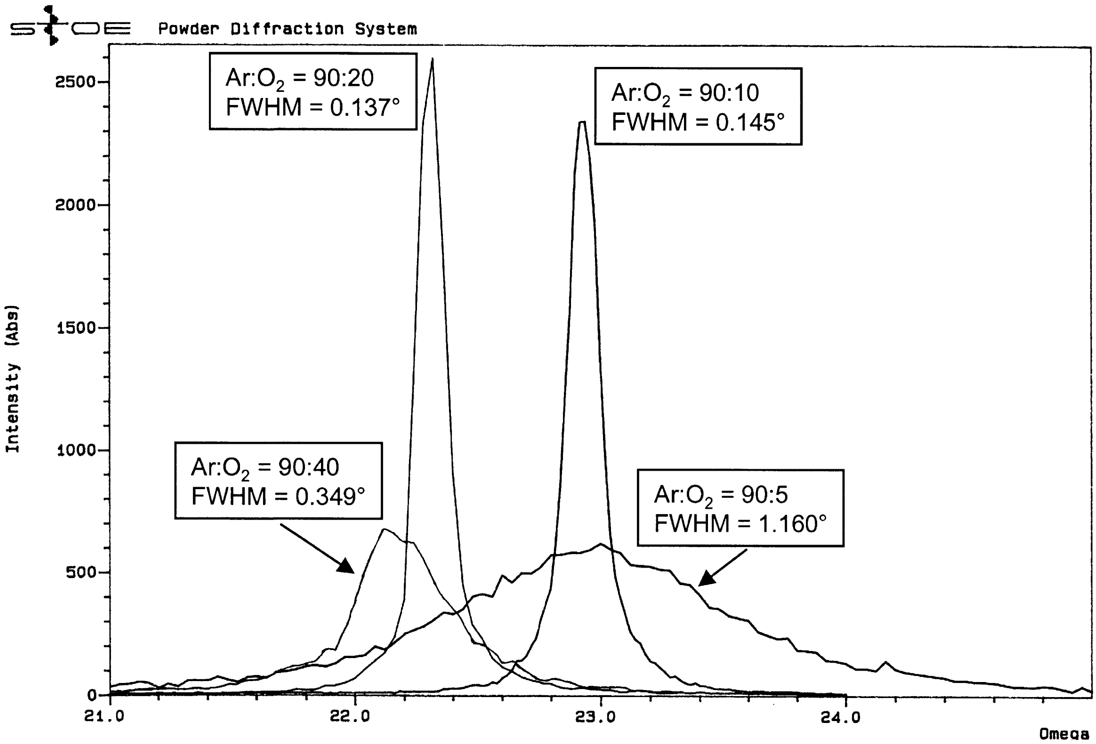

Fig. 3. ω-Scan pattern for the (200) reflection of BST thin films deposited using various sputtering gas ratio ($Ar:O_2$).

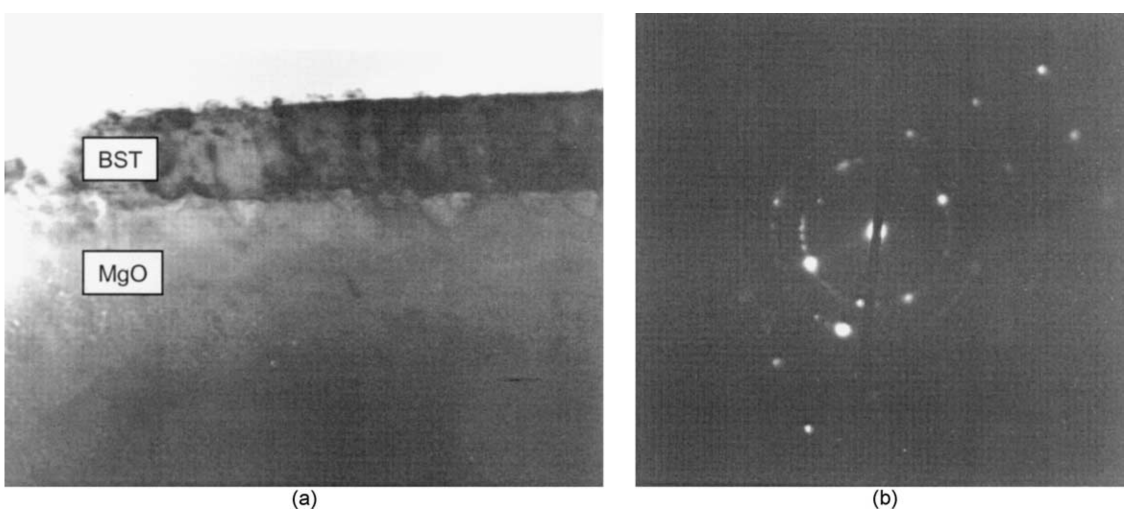

Fig. 4. (a) Cross-sectional TEM image and (b) SAD pattern of BST thin film grown on MgO substrate.

Table 2
Dielectric properties of various BST thin films measured at 1 MHz, RT

Sample	Preparation condition	ε_r@ 0 kV/mm	tanδ @ 0 kV/mm	Tunability [%] @ E_{max} [kV/mm]
BST/MgO	T_S = 600 °C	105	0.055	0.55 @ 15
BST/$LaAlO_3$	as deposited	234	0.020	0.66 @ 15
BST/MgO	T_S = 650 °C	125	0.0048	5.0 @ 15
BST/$LaAlO_3$	as deposited	206	0.0046	1.8 @ 15
BST/MgO	T_S = 700 °C	180	0.0037	16.2 @ 20
BST/$LaAlO_3$	as deposited	219	0.0036	2.2 @ 20
BST/MgO	T_S = 650 °C	287	0.0023	30.5 @ 15
BST/$LaAlO_3$	annealed in air: 900 °C/5 h	280	0.0046	10.3 @ 15

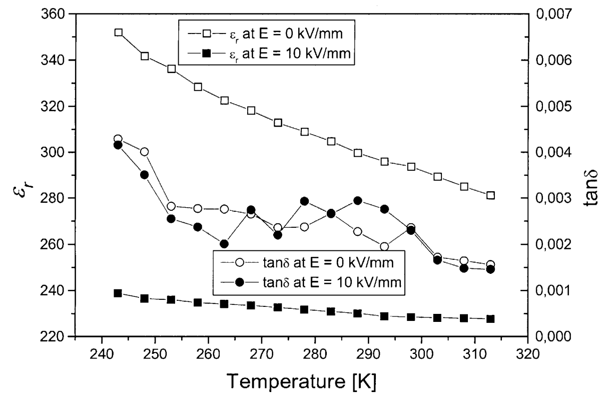

Fig. 5. Dependence of dielectric constant (ε_r) and loss tangent ($\tan\delta$) on temperature of optimized BST thin films measured at different electric fields at 1 MHz.

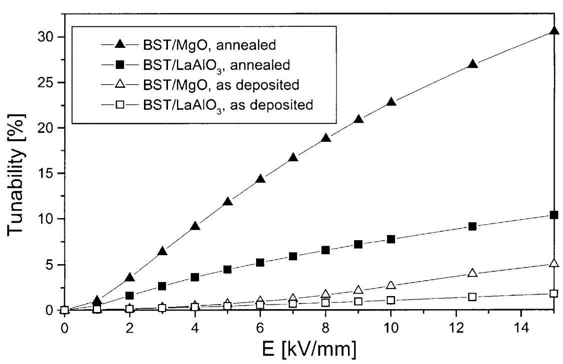

Fig. 6. Influence of substrates and post-annealing on tunability shown by electric field dependence, measured at 1 MHz, RT.

Table 3
Specific parameters of MgO and $LaAlO_3$ substrates in comparison with BST ($x = 0.5$)

Material	Crystal structure	Lattice Constant (Å)	Lattice mismatch to BST	Thermal expansion (K^{-1})
$LaAlO_3$	Quasi-cubic	3.789	−4.2%	$9.2 \cdot 10^{-6}$
MgO	Cubic	4.213	+6.3%	$12.8 \cdot 10^{-6}$
BST($x = 0.5$)	Cubic	3.947	/	$10.5 \cdot 10^{-6}$

4. Summary

The dielectric properties of RF sputtered BST thin films influenced by substrates and post-annealing were investigated. BST films deposited on MgO substrates under tensile stress showed higher tunability values than those deposited on $LaAlO_3$ substrates under compressive stress. Tunability can be significantly increased by post-annealing in air at 900 °C for 5 h. A tunability of 22% at $E_{max} = 10$ kV/mm and a loss tangent of 0.0023 have been achieved for an optimized sample measured at 1 MHz and room temperature.

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