

# Growth of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3/\text{LaAlO}_3$ Heterostructures by Injection MOCVD for Microwave Applications

J. Lindner,<sup>a\*</sup> F. Weiss,<sup>a</sup> J.-P. Sénateur,<sup>a</sup> V. Galindo,<sup>a</sup> W. Haessler,<sup>b</sup> M. Wehnacht,<sup>b</sup> J. Santiso<sup>c</sup> and A. Figueras<sup>c</sup>

<sup>a</sup>INPG-ENSPG-LMGP, UMR 5628 CNRS, St. Martin d'Hères, France

<sup>b</sup>IFW DRESDEN, Institut für Metallische Werkstoffe, Dresden, Germany

<sup>c</sup>ICMAB, CSIC, Bellaterra, Spain

## Abstract

It is well known that including dielectrics in layered structures such as  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3/\text{LaAlO}_3$  could be used as a basis for devices with voltage control of microwave circuit parameters. In this study,  $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  (BST) ( $x=0$  to 1) thin films have been epitaxially grown on  $\text{LaAlO}_3$  at a substrate temperature of  $800^\circ\text{C}$  using a new liquid source delivery technique called injection MOCVD. This process, based on computer-controlled injection of micro-amounts of liquid droplets, gives rise to BST thin films with their  $\langle 100 \rangle$  orientation perpendicular to the substrate displaying a FWHM of as low as  $0.14^\circ$  for the 002 diffraction  $\omega$  scan. AFM studies of the films, surface morphology revealed a smooth surface. In a next step dielectric properties are discussed. Finally, the possibility of obtaining  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3/\text{LaAlO}_3$  heterostructures was investigated, resulting in quite promising values for the critical temperature  $T_c$  of 88 K for the YBCO films. © 1999 Published by Elsevier Science Limited. All rights reserved

**Keywords:**  $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ , films, X-ray methods, electrical properties.

## 1 Introduction

High quality ferroelectric thin films offer unique opportunities for the development of advanced

microwave signal processing devices. In a ferroelectric the dielectric constant can be varied by applying an electric field. The variable dielectric constant results in change in the phase velocity in the device allowing it to be turned in real time for a particular application. The use of ferroelectric materials as a non-linear dielectric at microwave frequencies and the integration of tunable dielectrics with conductors that have low microwave surface ( $R_s$ ) is currently being investigated for a variety of advanced high frequency applications.<sup>1–3</sup>

We have investigated the growth of BST thin films ( $x=0–1$ ) and the deposition of bilayer structures for coplanar wave guides such as BST/YBCO heterostructures.

## 2 Experimental

$\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  (BST) ( $x=0$  to 1) thin films have been epitaxially grown on  $\text{LaAlO}_3$  at a substrate temperature of  $800^\circ\text{C}$  using a new liquid source delivery technique called injection MOCVD.<sup>4,5</sup>

Shortly, the precursor solution is contained in a hermetically closed vessel, pressurized under 1.25 bar of argon and connected to an injector (fuel injector used in recent thermal motors) (Fig. 1), which is a high speed electro-valve (Fig. 2). The droplets injected (precursor + solvent) into the evaporator, held at  $280^\circ\text{C}$ , are flash volatilized.

Growth conditions are described elsewhere.<sup>4</sup> Film thickness was determined using ellipsometry. Film composition was controlled by means of WDS, resulting in film composition of (Ba + Sr)/Ti between 0.95 and 1.05.

\*To whom correspondence should be addressed at LMGP-ENSPG, BP46, 38402 St. Martin d'Heres, France. Fax: +33-04-7682-6310; e-mail: lindner@adminpg.inpg.fr

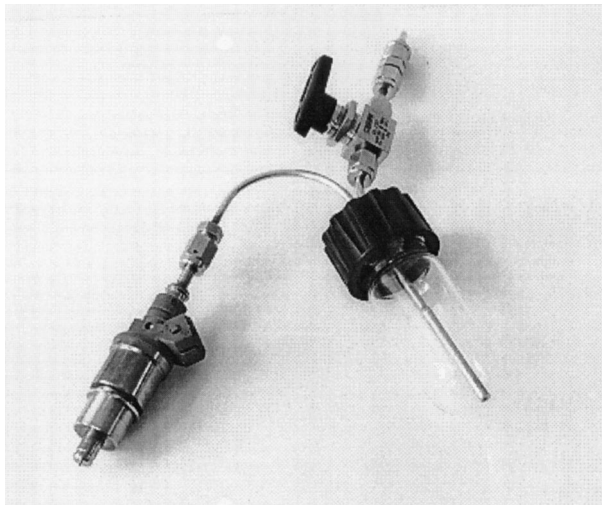


Fig. 1. Injector and hermetically closed vessel.

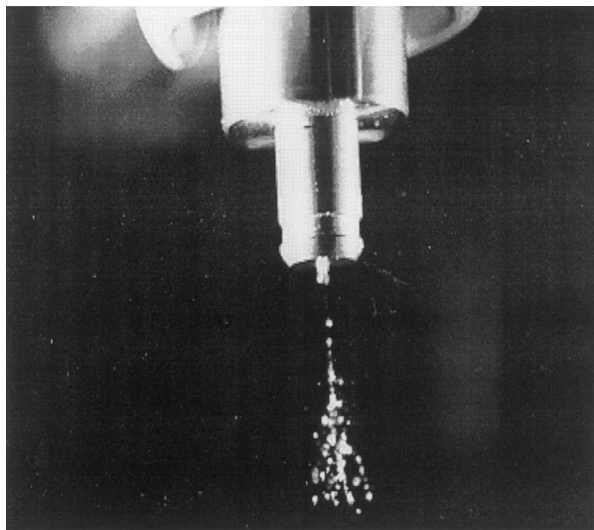


Fig. 2. Injected droplet.

### 3 Results

$\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  ( $x=0-1$ ) films were deposited by injection MOCVD onto  $\text{LaAlO}_3$  substrates. The 1000 Å thick films were characterized by X-ray diffraction. In Fig. 3 one can see the diffraction pattern for a single phase STO film deposited onto  $\text{LaAlO}_3$ .

Rocking curves for the BST films ranged from  $0.14^\circ$  ( $x=0$ ) to  $0.45^\circ$  ( $x=1$ ) for the (200) reflection (Fig. 4). In fact, the values for FWHM increase steadily with increasing  $x$ , resulting from an increasing lattice mismatch between substrate and  $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  film.

Furthermore in-plane epitaxy was studied by performing  $\Phi$ -scans of the (101) $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  film diffraction. One peak every  $90^\circ$  clearly shows the epitaxial growth of  $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  film on the

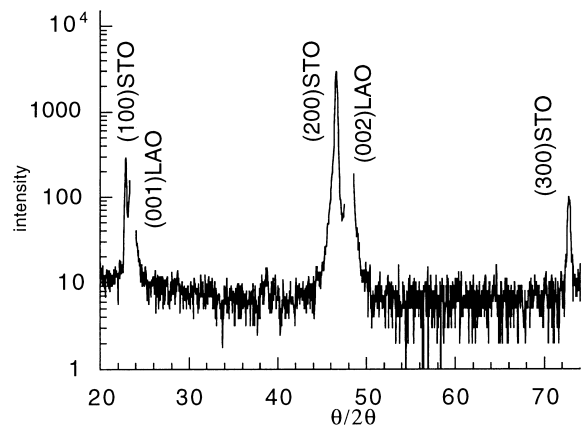


Fig. 3. X-ray pattern of  $\text{SrTiO}_3$  (STO) film.

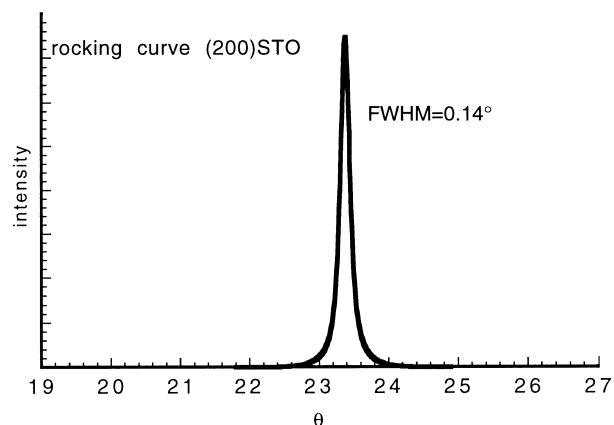


Fig. 4. Rocking curve of  $\text{SrTiO}_3$  (STO) layer.

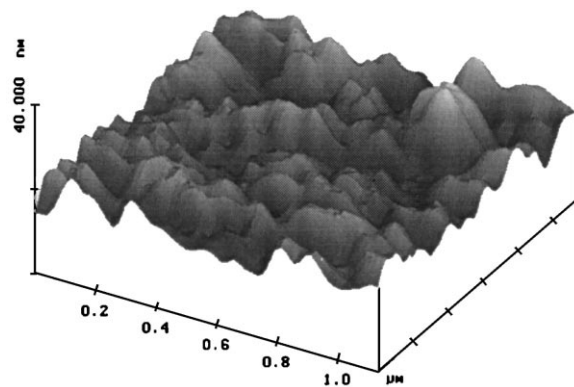


Fig. 5. AFM scan of  $\text{BaTiO}_3$  film ( $R_s=3$  nm).

$\text{LaAlO}_3$  substrate. The full width at half maximum of the peaks yields values  $\text{FWHM}=0.7^\circ$  and  $0.3^\circ$  for the  $\text{SrTiO}_3$  film and the  $\text{LaAlO}_3$  substrate, respectively. The coincidence of the maxima of the  $\Phi$ -scans for the deposited film and the substrate shows that the growth of  $\text{SrTiO}_3$  on  $\text{LaAlO}_3$  substrates is characterized by an epitaxial cube-on-cube relationship.

An AFM study revealed a quite flat surface with a surface roughness varying from 1 nm for  $\text{SrTiO}_3$  to 3 nm in the case of  $\text{BaTiO}_3$  (Figs 5 and 6).

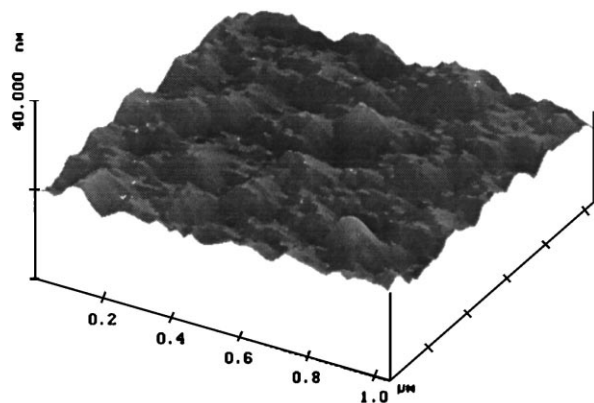


Fig. 6. AFM scan of  $SrTiO_3$  film ( $R_s = 1$  nm).

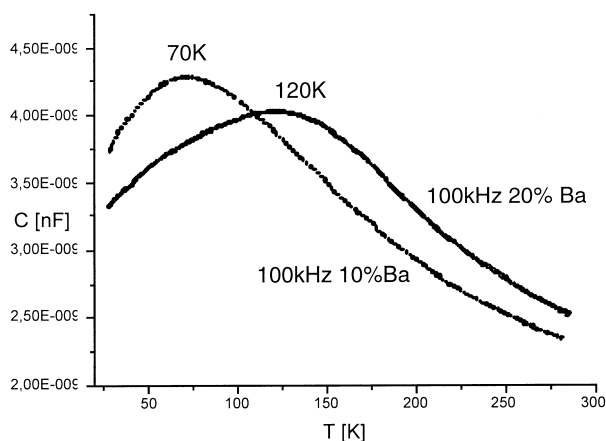


Fig. 7. Capacitance measurements of BST films.

In a next step, the temperature dependence of the dielectric properties, crucial for the microwave tunability, was studied using  $Ba_xSr_{1-x}TiO_3$  films ( $x=0.1$  and  $0.2$ ) deposited on conducting Nb doped  $SrTiO_3$  single crystals [ $SrTiO_3:Nb$ , (100 orientation)] which can be used as electrodes (Fig. 7).

The maxima of the dielectric constant were found to be at the same temperature value as for bulk crystals. Examining an eventual shift of these maxima positions, the same kind of capacitance measurement will be done in near future for  $Ba_xSr_{1-x}TiO_3$  films, deposited on  $LaAlO_3$ , using interdigital electrodes on the surface of the  $Ba_xSr_{1-x}TiO_3$  films.

Finally, investigating the deposition of bilayers structures, YBCO thin films were deposited on the  $Ba_xSr_{1-x}TiO_3$  ( $x=0.1$ ) layers at a temperature of

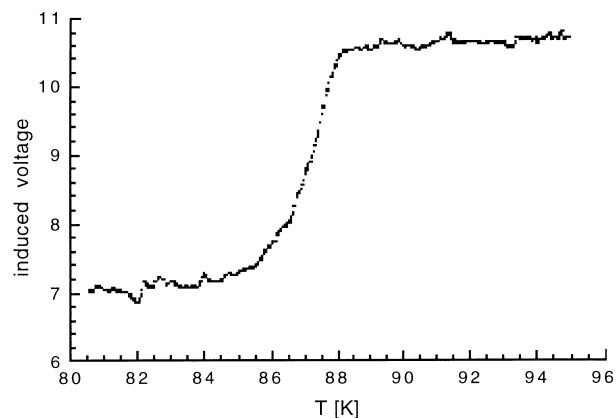


Fig. 8.  $T_c$ -measurement.

850°C. The resulting thin layers displayed a critical temperature of  $T_c = 88$  K (Fig. 8) for the YBCO layer.

#### 4 Conclusion

High quality  $Ba_xSr_{1-x}TiO_3$  thin films have been deposited by injection MOCVD onto single crystal substrates of  $LaAlO_3$ . The deposited films, oriented both with respect to the substrate surface normal and in the plan of the films, display rocking curve width for the (200) reflection of as low as  $0.14^\circ$ .

As for the superconducting YBCO films deposited onto the BST layers, the preliminary results are very promising ( $T_c = 88$  K).

#### Acknowledgement

This work is supported by the European Community under contract number IC15-CT96-0735.

#### References

1. EUCAS'97 Proceedings, Eindhoven, The Netherlands, 1997.
2. Vendik, O. G. *et al.*, *Ferroelectrics*, 1993, **144**, 33-43.
3. de Groot, D. C. *et al.*, *IEEE, Transactions on Applied Superconductivity*, 1997, **5**(2), 2272.
4. Felten, F., Sénateur, J. P., Weiss, F., Madar, R., Abrutis, A., *Journal de Physique IV, Colloque C5, Journal de physique II*, 1995, **5**, C5-1079.
5. Sénateur, J. P., Weiss, F., Madar, R., Abrutis, A., Patent 93/08838 (France), 1993.