Strain at the nanoscale in epitaxial $BaTiO_3$ films on silicon

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1. INTRODUCTION

Integration of ferroelectrics on semiconductors would offer the opportunity to add novel functionalities on chips (logic, memory, sensors…) [1, 2]. For such a purpose, molecular beam epitaxy (MBE) provides unique advantages to precisely construct the oxide/semiconductor interface, which plays a major role in nanoelectronic devices. However, the direct epitaxy on silicon of a perovskite oxide such as $BaTiO_3$ is challenging due to the oxidation of the silicon surface and due to the large lattice mismatch and thermal expansion mismatch between the oxide and the semiconductor. One solution is interface engineering using Sr- passivation and epitaxial growth of $SrTiO_3$ templates on Si substrates. Then subsequent epitaxial growth of $BaTiO_3$ can be explored.

In this study, a quantitative analysis of high-resolution transmission electron microscopy (HR(S)TEM) images using the geometric phase analysis (GPA) [3] is proposed in order to support the growth strategy of epitaxial $BaTiO_3$ films with the desired orientation, i.e. with the c-axis of the tetragonal structure perpendicular to the Si substrate.

2. RESULTS

2.1 Experimental

Several epitaxial stacks were grown by MBE. First a 4 nm-thick epitaxial $SrTiO_3$ buffer layer was grown on HF-cleaned p-type Si (001) substrate. The subsequent growth of $BaTiO_3$ was realized by co-evaporating Ba and Ti at a temperature ranging typically from 440 °C to 525 °C and an oxygen partial pressure of typically 1 to $5x10^{-7}$ Torr. The temperature, oxygen pressure, and cooling down conditions were varied to optimize the growth conditions [4]. An oxygen plasma source could be used for post-deposition anneal of the films.

With GPA, maps of the strain in the $BaTiO_3$ films with respect to the Si substrate are determined with a high precision (0.1%) at the nanometric scale (1-2nm). From these maps, the local lattice parameters and thus the tetragonality (c/a ratio) of the $BaTiO_3$ films can be evidenced [5]. HRTEM work is performed on an image corrected Hitachi HF3300S microscope (I2TEM-Toulouse) and HR(S)TEM on a FEI Titan Low-Base 60-300 (Zaragoza).

2.2 Strain maps and tetragonality

Atomic structure images of the stacks evidence that during the $SrTiO_3$ and $BaTiO_3$ growth and anneals, oxygen diffusion occurs through the film leading to the formation of an interfacial amorphous SiO_2 layer between the Si substrate and the $SrTiO_3$ buffer layer, but preserving the original epitaxial relationship (Figure 1).

Typical in-plane and out-of-plane strain maps in the STO/BTO stack are illustrated on figure 2b and c. They result from the GPA analysis of the associated HAADF image (Figure 2a). From these maps, the local lattice parameters and thus the tetragonality in the BTO film is determined. The corresponding profiles obtained by integration over a defined area are illustrated on figure 2 d and e respectively.

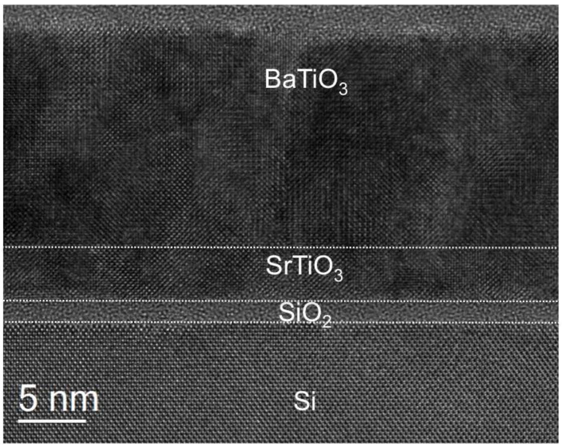

Figure 1. HRTEM (I2TEM) image of the $Si/SiO_2/SrTiO_3/BaTiO_3$ stack.

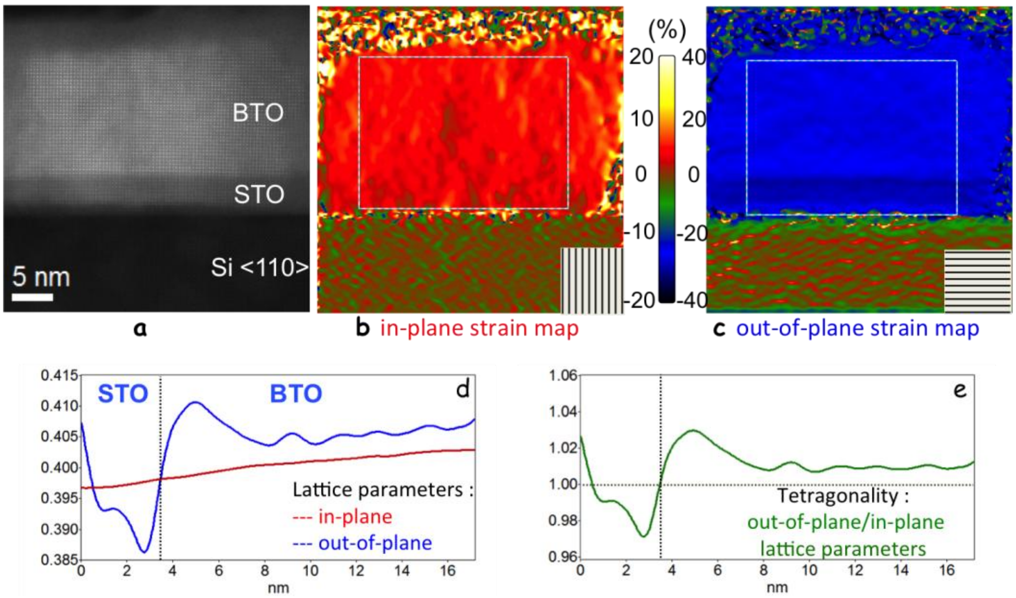

Figure 2. HAADF image of a $Si/SiO_2/SrTiO_3/BaTiO_3$ stack (a), corresponding strain maps within the STO/BTO stack (b, c) and extracted profiles of the lattice parameters within the stack (d) and the associated tetragonality (e).

3. CONCLUSION

BTO films exhibit mainly a c-axis growth with a gradual change of the in-plane lattice parameter and stronger change of the out-of plane parameter. The tetragonality is always maximum just above the STO/BTO interface and stabilizes after few nanometers above this interface. Correlation with EELS analyses of cationic and O compositions will be proposed.

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