Ferroelectric oxides integrated on a semiconductor substrate are of particular interest for various applications such as memory or logic devices\(^1\,^2\). Among the ferroelectric materials, BaTiO\(_3\) is an attractive candidate for large-scale applications compared to Pb- or Bi-based compounds. It is a well-known perovskite largely studied for its dielectric, piezoelectric and ferroelectric properties. However, the direct epitaxy of BaTiO\(_3\) on silicon is challenging due to the oxidation of the silicon surface and due to the large lattice mismatch (4\%) and thermal expansion mismatch between the oxide and the semiconductor. Moreover, the control of the ferroelectric polarization is a crucial topic for the targeted applications. The polarization must point perpendicular to the Si channel.

In this study, a quantitative analysis of high-resolution transmission electron microscopy (HRTEM) 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. 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). Strain maps with improved precision (0.05\%), 5 nm spatial resolution and with a large field of view (1 \(\mu\)m) are also proposed for selected samples, using dark field electron holography\(^4\). From these maps, the local lattice parameters and thus the tetragonality (c/a ratio) of the BaTiO\(_3\) films can be evidenced\(^5\). The HRTEM images and holograms were acquired on a Hitachi HF3300S microscope (I2TEM-Toulouse) fitted with the new aplanatic spherical aberration corrector B-COR from CEOS and a 4096x4096 camera.

Growth of the epitaxial BaTiO\(_3\) films was performed by molecular beam epitaxy (MBE) using an SrTiO\(_3\) epitaxial buffer layer to reduce both thermal and lattice mismatches on Si (001). Different process parameters like the growth temperature and cooling conditions were explored to optimize the quality of 20 nm thick BaTiO\(_3\) films and to minimize the SiO\(_2\) interfacial layer regrowth between Si and the SrTiO\(_3\) buffer. The influence of the thickness of the SrTiO\(_3\) buffer layer on the growth mode of 10 nm thick films was examined. Finally, in order to investigate the first steps of the BaTiO\(_3\) formation, the growth behavior of thin films with thicknesses ranging from 1.2 to 4 nm (3 to 10 monolayers) was considered.

1. J. Scott, Ferroelectric memories (Berlin: Springer), chapter 2 and 12 (2000)
5. C. Dubourdieu et al., Nature Nanotechnology 8, 748 (2013)

Acknowledgement: This work has been supported by the French National Research Agency under the reference No. ANR-10-EQPX-38-01. The authors acknowledge the "Conseil Regional Midi-Pyrénées" and the European FEDER for financial support within the CPER program.
Fig. 1: HRTEM image of a MBE grown BaTiO$_3$ film on a Si substrate using an SrTiO$_3$ epitaxial buffer layer; Strain maps in the BaTiO$_3$ film along two perpendicular directions; Corresponding ratio profile of the retrieved BaTiO$_3$ lattice parameters revealing the tetragonality of the film with the largest parameter (c axis) being perpendicular to the substrate.

Ratio of the BaTiO$_3$ lattice parameters retrieved from the above strain maps as a function of the distance (nm) from the SiO$_2$/SrTiO$_3$ interface along the BaTiO$_3$ growth axis direction (y)