Investigation of growth of thin layers of perovskite on native silicon dioxide by a combination of atomic force microscopy and transmission electron microscopy

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Abstract. Thin layers of (Sr,Ba)TiO_3 perovskite have been grown on native silicon dioxide by pulsed laser deposition at the Technical University of Darmstadt, Germany. Atomic force microscopy (AFM) has been used to investigate the surfaces of the native silicon oxide before and after over-growth by the perovskite in plan-view. Bright-field and dark-field scanning transmission electron microscopy (STEM) in a JEOL 2010F field-emission transmission electron microscope have been combined to investigate the layer stacks of Si/SiO_2/(Ba,Sr)TiO_3 in cross-section. The aim is to correlate surface roughnesses in plan-view geometry with interface roughness in cross-sectional geometry, with an emphasis on detecting percolation in the perovskite layers if they approach thicknesses of only a few unit cells.

1. Introduction

Many III-V compound semiconductors such as GaAs, InAs and InSb, have higher electron mobility than Si and also a direct band-gap which makes them more attractive for use in electronic and opto-electronic applications. However, silicon is mainly used in semiconductor devices due to its chemical stability, thermal properties, scalable processing of its oxide and cost. Hence III-V epitaxy on Si has been studied for years.

Because of the mismatch in lattice parameters between different semiconductors (other than AlAs/GaAs) there usually is a high density of dislocations at the interface. One solution to avoid this is to use an intermediate layer between silicon and III-V semiconductor. One choice for the intermediate layer would be a cubic perovskite-type oxide. All perovskites have lattice constants near 4Å which is close to $\sqrt{2}$ times the lattice constant of GaAs (≈ 5.65Å); therefore GaAs could potentially be grown on the perovskite layer by 45° in-plane rotation which could give fewer dislocations compared to direct growth of GaAs on Si [1]. In this work we investigate the growth of thin layers of perovskite on native silicon dioxide before overgrowth by GaAs.

2. Experimental details

Four wafers, each consisting of a thin layer of perovskite grown on Si substrates via the native oxide by pulsed laser deposition, have been analyzed by atomic force microscopy (AFM), ellipsometry and scanning transmission electron microscopy (STEM). Two of the samples have thin layers of $SrTiO_3$

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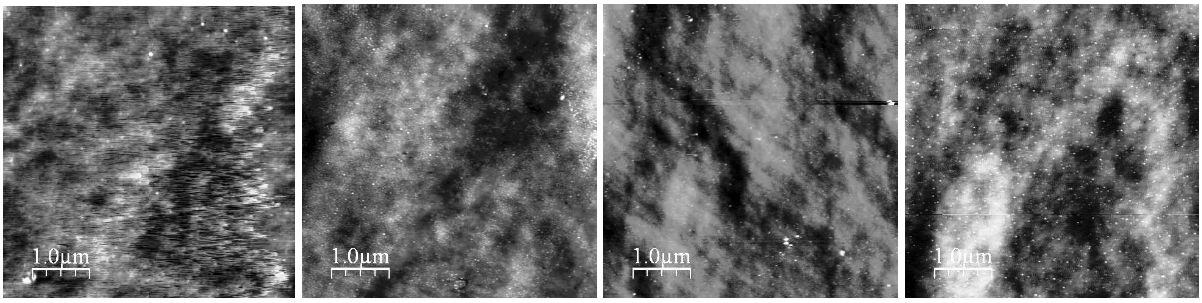
with nominal thicknesses of 2 and 8nm and the other two have layers of $BaTiO_3$ of same nominal thicknesses.

AFM images have been captured by a Veeco Nanoscope V instrument operated in tapping (non-contact) mode. Nanotec's WSxM 5.3 software [2] was used to analyse the AFM images. A VASE® J.A. Woollam Co., Inc. ellipsometer has been used to measure the thickness of the perovskite layers. To investigate the surface of thin perovskite layers and the interfaces between Si and the layers by electron microscopy, cross-sectional samples were prepared by gluing two pieces of wafers, one with nominally 2nm perovskite layer and one with nominally 8nm perovskite layer, face to face, gluing Si backing blocks to each side to make them 3mm thick, cutting the blocks by a diamond saw, grinding and polishing the cross-sectional samples and finally ion milling them in a Fischione 1050 argon ion mill at 4kV and 2mA until electron transparency. The electron microscope used in this study is a JEOL-2010F field-emission transmission electron microscope operated at 197kV, which has a scan unit and bright-field (BF) and annular dark-field (ADF) detectors for scanning transmission electron microscopy (STEM). The microscope is also equipped with a GATAN imaging filter with a charge coupled device (CCD) camera for image acquisition.

3. Results

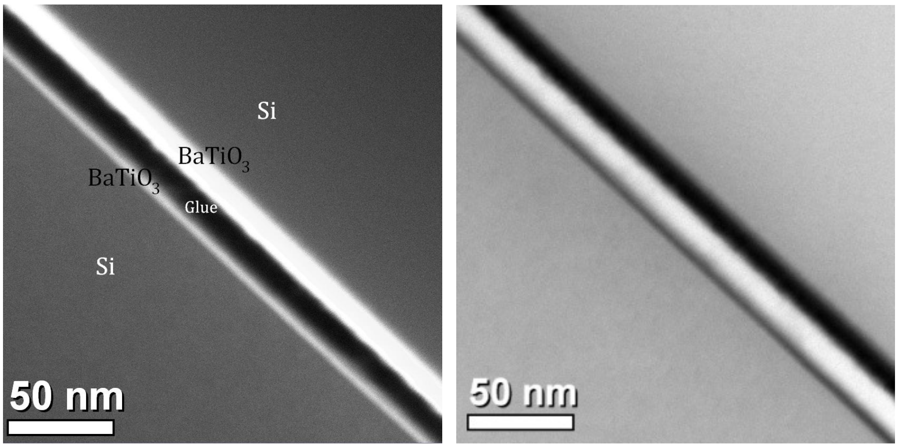
Figure 1 shows the AFM images of areas of 5×5 μm^2 for each wafer with a thin layer of $BaTiO_3$ (a: 2nm, b: 8nm) or $SrTiO_3$ (c: 2nm, d: 8nm nominal thickness). The acquisition time was 1s for all images which consist of 512 × 512 pixels (yielding a sampling of 9.8nm/pixel).



(a) **(b)** **(c)** **(d)**

Figure 1. AFM images of 5μm × 5μm **(a)** $BaTiO_3$ (2nm) **(b)** $BaTiO_3$ (8nm) **(c)** $SrTiO_3$ (2nm) and **(d)** $SrTiO_3$ (8nm). The image contrast of black to white corresponds to a height of 10nm in all images.

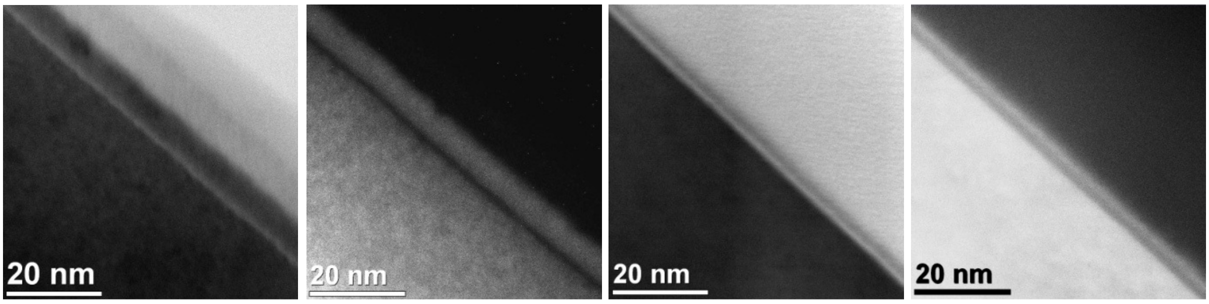
Figures 2a and 2b are annular dark-field (ADF) and bright field (BF) STEM images of the cross-sectional sample of $BaTiO_3$ acquired with spot size of 0.3 nm and dwell time of 19.5μs. The beam convergence semi-angle is equal to 9.5mrad, the collection angles are equal to 0-10mrad for BF and 55-170mrad for ADF. The differences of contrast along the layer in bright-field confirm that the perovskite layer is not epitaxially grown but poly-crystalline. The field of view of 844nm is sampled onto 4096 × 4096 pixels at a sampling rate of 0.21nm/pixel.



(a) **(b)**

Figure 2. Central regions of **(a)** ADF and **(b)** BF-STEM images of cross-sectional sample of $BaTiO_3$ (2 and 8nm titanate layers appear bright in ADF and dark in BF and show surface roughness).

The corresponding BF and ADF-STEM images of cross-sectional samples of $SrTiO_3$ (2 and 8nm) are shown in figure 3. Again in bright-field, lateral fluctuations in contrast along the $SrTiO_3$ layers can be observed which indicates the layers are poly-crystalline. The field of view of 124nm was scanned with 2048 × 2048 pixels at a dwell time of 19.5μs, which gives a sampling equal to 0.06nm/pixel.



(a) **(b)** **(c)** **(d)**

Figure 3. Central parts of **(a,b)** BF and ADF-STEM images of 8nm $SrTiO_3$ and **(c,d)** BF and ADF-STEM images of 2nm $SrTiO_3$.

To obtain the roughness from the STEM images, first the images have been converted to binary format using thresholding, and then a MATLAB code was applied to each image column to work out where the transition from 0 to 1 occurs, which locates the surface of the perovskite layer. Knowing these points for all image columns and calculating a straight line that best fits the data, the difference between the real data and the linear estimate describes the roughness of the surface of the layers. The standard deviation of this data is the root-mean-square (RMS) interface roughness in projection. Figure 4 shows the roughness data acquired from the BF-STEM image shown in Figure 2b for the 8nm BTO as an example.

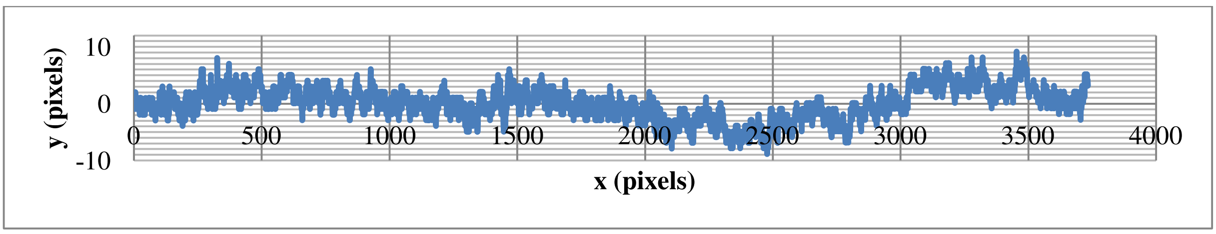


Figure 4. The projected surface roughness acquired from the BF-STEM image of 8nm $BaTiO_3$

Table 1 shows the RMS surface roughness of all four samples measured and calculated from both AFM and STEM images.

Table1. RMS roughness in nm, measured from the AFM images and the STEM images

	$BaTiO_3$(8nm)	$BaTiO_3$(2nm)	$SrTiO_3$(8nm)	$SrTiO_3$(2nm)
AFM	0.61	0.48	0.57	0.55
STEM	0.58	1.09	0.53	0.73

The surface roughness measured from AFM images and STEM images don't agree with each other for very thin layers. As STEM measures the interface roughness as projected along the sample thickness, small protrusions or indentations will be smeared out, therefore the roughness value from STEM images should be always smaller than the true values. Similarly, for very thin layers (2nm) the grains are probably narrower than the tip radius and also smaller than the sampling, and integration over several grains will also reduce the apparent surface roughness.

Using an ellipsometer the thicknesses of perovskite layers for all four wafers have been measured. Table 2 compares the thicknesses of the perovskite layers measured by ellipsometry and STEM.

Table2. Thickness of perovskite layer in nm, measured by ellipsometry and from the STEM images

	$BaTiO_3$(8nm)	$BaTiO_3$(2nm)	$SrTiO_3$(8nm)	$SrTiO_3$(2nm)
STEM	10.32	2.68	5.92	1.82
ellipsometry	9.22	2.31	5.59	2.39

The thicknesses of the perovskite layers measured by ellipsometry roughly agree with the measurements by STEM. Thicknesses of both $BaTiO_3$ layers and the nominally 8nm $SrTiO_3$ layer are ~10% larger than ellipsometry suggests, which may be due to uncertainties in the complex dielectric function of the perovskite.

4. Conclusion

The thicknesses and surface roughnesses of thin layers of poly-crystalline (Sr, Ba)TiO_3 grown on silicon substrate via native oxide have been investigated using atomic force microscopy, ellipsometry and scanning transmission electron microscopy. The roughnesses and thickness measured by different methods have been compared. From our data it appears likely that the thinnest perovskite layers of ~2nm thickness have an RMS roughness of at least 0.7-1.1 nm, which is ⅓ - ½ of the layer thickness and indicates that the layers will probably not be completely continuous and might have pin-holes

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