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Study of roughness evolution and layer stacking faults in short-period atomic layer deposited HfO₂/Al₂O₃ multilayers

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In this work we study the evolution of roughness in interfaces of HfO_2/Al_2O_3 multilayers by x-ray reflectivity. It was found that, besides the reduced adatom surface mobility during atomic layer deposition, an improvement of the interface quality can be achieved upon the stacking of several layers. Although the low roughness of the initial surface could not be recovered, there was a considerable improvement of surface/interface quality along the deposition process. In particular, variations on the growth temperature were not able to tailor the surface quality, if compared to the stacking process. Finally, transmission electron microscopy analysis has shown that local defects can take place among nearly perfect interfaces. Such effect must be taken into account for nanometer-scale device fabrication. © 2011 American Institute of Physics. [doi:10.1063/1.3555624]

I. INTRODUCTION

High dielectric constant (high- κ) materials have been intensively studied over the past few years due to their crucial relevance for novel electronic device development. Many new complementary metal-oxide-semiconductor or compact capacitor architectures can be achieved by employing high- κ oxides.^{1,2} By using high- κ materials it is possible to make use of ultra-thin oxide layers for the gate barrier in transistors.³ Atomic layer deposition (ALD) has become the standard growth method for obtaining high- κ oxides with controlled thickness. Key advantages of the method are conformal coverage, low contamination of the deposited material and layer-by-layer deposition on the atomic layer scale.⁴

Metallic oxides such as Al_2O_3 and HfO_2 exhibit high dielectric constants and large energy gaps and are suitable for electronic applications. In spite of its high- κ properties, the efficiency of thin HfO_2 layers to insulate is strongly affected by annealing at large temperatures (T > 500 °C).⁵ Such phenomenon takes place due to crystallization, that leads to the reorganization of atoms and the formation of defects. These defects may act as preferential channels for current leaks on a device, compromising its usage.⁶ In such case a simple solution to this deficiency is found by combining different oxide layers. Stacking HfO_2 and Al_2O_3 layers, for instance, can lead to improved thermal stability for devices since alumina remains amorphous up to ~1000 °C.^{7,8} Besides electric conductivity properties, thermal conductivity is also affected by increasing the number of interfaces in a layered system.⁹ Such procedure affects the propagation of phonons that will scatter at the interfaces depending on the differences in elastic properties and densities of vibrational states of the successive layers. Therefore, the number of interfaces – and in some cases the interface roughness quality – are key parameters to tune macroscopic properties.

The relation between layer stacking and interface quality in thin films has been analyzed for different growth methods, such as thermal deposition and sputtering.¹⁰ However, roughness evolution in ALD multilayers still needs further clarification. In the 10–40 Å scale limit, the layer thickness in such stacks approaches the native roughness of the best substrates used for applications. In spite of having a conformal coverage process, the interface roughness may worsen as layers stack, becoming as large as the layer thickness itself.

In this work we have used x-ray reflectivity (XRR) and transmission electron microscopy (TEM) to evaluate interface roughness in a series of HfO₂/Al₂O₃ multilayers. The total of stacked layers and the growth temperature were varied, allowing for understanding variations due to coverage and surface adsorption. The results have shown that roughness decreases with increasing number of stacked layers, but is mainly unaffected by deposition temperature. TEM images show that localized defects can be formed during deposition, in contrast to the statistically averaged XRR results.

II. EXPERIMENTAL

In order to investigate the evolution of roughness and interface quality in ALD multilayers, a complete set of samples was grown on top of Si(001) substrates with native

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SiO₂. The native SiO₂ thickness was expected to range in between 15 and 25 Å, and the average roughness of the starting surface was measured by atomic force microscopy (AFM), yielding a value of 2 Å. The ALD oxide deposition took place in a Savannah 100 reactor, where the sample is heated from its back. The carrier gas (N_2) had a fixed flow rate of 20 sccm along the whole process. Trimethyl-aluminum and water, both at room temperature, were used as precursors for Al₂O₃. HfO₂ layers were formed by using tetrakis(dimethylamino)hafnium and water, with the Hf precursor heated at 75 °C. The pulse duration and intervals for all precursors were identical: 0.015 and 30 s, respectively. Such conditions yielded a growth rate of 0.9 Å/cycle for Al_2O_3 and 1 Å/cycle for HfO₂. The total layer thickness of all samples were found to vary within $\sim 2\%$ among the different growth temperatures used, indicating that growth rates remain unchanged in the temperature range explored in this work (see discussions in Sec. IV).

A general layout for multilayer stacks used in this work is shown in Fig. 1(a). The deposition was performed alternating 20 Å Al₂O₃/20 Å HfO₂ layers (nominal thickness), with distinct total stack number (n). In the first growth series, hereafter called series A, the substrate temperature was fixed to 140 °C, and n ranged from 2 to 40, with samples at n = 4, n = 8, and n = 20. In the second series – referred as series B–n was fixed to 8, while substrate temperatures of 110, 140, and 180°C were used during growth. From such parameters, the A series was designed to investigate the evolution of interface properties with respect to the total amount of layers deposited. In the B series, the temperature variation was used to modify the



FIG. 1. (Color online) (a) Layer stack layout for ALD deposition in both grown sample series. (b) XRR measurements for the n=8 ($T_g=140$ °C) sample (dots), with fits using the kinematical model (solid line in high-q region) and the Parratt model (solid line in low-q region).

precursor surface kinetics, allowing to infer changes in the interface quality due to growth conditions.

X-ray reflectivity (XRR) measurements were performed at the XRD1 beamline of the Brazilian Synchrotron Light Laboratory (LNLS). In this beamline, a monochromatic xray beam is obtained from a Si(111) double crystal monochromator. The energy was fixed to 8 keV, and a 220 μ m collimated beam at the vertical direction (scattering plane) was employed. For the XRR measurements the long axis of the studied samples (2 cm) were aligned along the beam direction, allowing for easier illumination corrections.¹¹ In our setup, the specular reflectivity signal was measured up to $2\theta = 8^\circ$, where the intensity reaches a constant background level.

TEM analyses were performed in a JEM3010-UHR microscope, working at 300 keV. Only the n = 40 sample was submitted to TEM evaluation, which was performed in an ion milled cross-sectioned sample. The interface evolution along the 40 Al₂O₃/HfO₂ bilayer repetitions was regarded as a systematic representative of the layer stacking for sample series A.

III. X-RAY REFLECTIVITY MODEL

For the layer stacks used here, the sample with maximum thickness used correspond to n = 40, where a total of 1600 Å of material was deposited. For such conditions, a kinematic model for XRR was employed in order to describe the main features of the measured data. Such approach has the advantage of being more sensitive to electronic density variations¹² and allows for obtaining correct parameters of thickness, electronic density and roughness, with a direct visualization of the influence of each of these parameters in the curve fitting.

The kinematic reflectivity model takes into account the difference in electronic density among layers. Considering the thickness (d_j), roughness (σ_j), and electronic density (ρ_j) for the j *th* layer, the general equation describing the reflectivity from a multilayer stack can be expressed as¹²

$$I\infty |r\phi(q)|^2 = r^2 \left| \sum_{j=1}^{N} (\rho_j - \rho_{j+1}) e^{-iQ\Sigma_j d_j} e^{-(Q\sigma_j)^2/2} \right|^2, \quad (1)$$

where the summation is performed over the total of layers N in each sample (N = 2n + 1, considering the SiO₂ layer). The Fresnel reflectivity of a bare substrate is taken into consideration by the term r.¹³ A typical result from such kinematical model is shown by the solid red line in Fig. 1(b), where a XRR measurement performed at the n = 8 sample is shown (dots).

The kinematical model provides a good fitting at high scattering angles. However, since it lies in Born approximation theory, it deviates considerably from the experimental profile close to the critical angle of total external reflection.^{12,13} In order to fit this low angle region of the curve, the Parratt recursive model with the Nevot-Croce approximation¹⁴ for roughness has been used. In this case, the structural parameters were not fitted but were taken exactly from the kinematical approach. Since the Nevot-Croce correction to

the Fresnel coefficient works well for $q_z \sigma < 1$ (Ref. 15) and according to our kinematical fitting $\sigma = 5-10$ A, there is a small region of overlap around $q_z = 0.1$ A⁻¹ where the two approximations match. The good matching between the different approximations and the experimental data shows the reliability of the fitting performed using kinematical approach. At large q_z values the Parratt model cannot be used to fit the results due to the high layer roughness/thickness ratio. The complete description of the Parratt model can be found in Refs. 12 and 16.

IV. RESULTS AND DISCUSSION

In order to obtain structural parameters from the XRR measurements, the analysis of reflectivity profiles from samples in series A and B was initially performed using the kinematical model. In our fits, the roughness of both HfO_2/Al_2O_3 and Al_2O_3/HfO_2 interfaces was considered to be the same for all interfaces along the whole stack. Similarly, the electronic density and thickness for all HfO_2 and Al_2O_3 layers was kept fixed for each compound in our system. Such constraints, allowed for a considerable reduction of the free fitting parameters, providing average information for the multilayer. Although this information cannot be regarded as locally correct for each interface/layer, it allows to understand deviations in the behavior of the ensemble of interfaces as well as to find out whether the system evolved into nonhomogeneous configurations.

In Fig. 1(b) the XRR data of the n = 8 sample for series A is shown (dots) with the corresponding best kinematical fit (low q_z region). Some information can be directly obtained from the measured data. The short period oscillations, that account for the total thickness of the multilayer are spaced by $\Delta q_z \sim 0.0177 \text{ Å}^{-1}$, leading to a total thickness $D = (2\pi/\Delta q_z) \sim 355 \text{ Å}$. Such value is compatible with the sum of nominal average 20 Å-thick Al₂O₃ or HfO₂ layers that form the multilayer stack, sitting on top of a ~20 Å thick native SiO₂ layer. Besides the short period oscillations, satellite peaks are observed at specific q_z positions, which represent constructive interference among all Al₂O₃/HfO₂ bilayers. From the position of the first satellite and the difference Δq_z from successive satellites, one obtains an average bilayer thickness.

From the kinematical fit to the XRR profile, consistent information of the layer thickness, roughness and electronic density is extracted. In the case of Fig. 1(b), the layer thicknesses were found to be $d_{A12O3} = 21.6$ Å, $d_{HfO2} = 20.1$ Å, and the average multilayer interface roughness could be evaluated as $\sigma_{A1Hf} = 5.9$ Å. Table I summarizes all fitting parameters for the samples of series A. Using the parameters obtained via kinematical model into the Parrat model, we were able to capture the behavior of the low-q_z region of the curve, proving the robustness of our approach.

For the analysis of all samples in both series A and B, the electronic densities were kept fixed, with values $\rho_{Si} = 0.699$ e⁻/Å³, $\rho_{SiO2} = 0.648$ e⁻/Å³, $\rho_{Al2O3} = 0.738$ e⁻/Å³, and $\rho_{HfO2} = 2.740$ e⁻/Å³, for the Si substrate, SiO₂ native layer, Al₂O₃ and HfO₂ layers, respectively. In all fits the roughness of the SiO₂/Al₂O₃ interface was fixed at 2 Å, matching the

TABLE I. Thickness and roughness parameters obtained from XRR analysis of the multilayers from the sample series A, varying the number of HfO₂/Al₂O₃ bilayers for a fixed temperature $T_g = 140^{\circ}$ C. The error bars for SiO₂ layer thickness in which a star is shown (*) cannot be determined since the multilayer is much thicker than the SiO₂ layer.

| Number of bilayers | $d_{Al2O3} {\rm \AA}$ | $d_{HfO2} {\rm \AA}$ | $d_{SiO2} {\rm \AA}$ | $\sigma_{\rm AlHf}{\rm \AA}$ | $\sigma_{ m HfAr}{ m \AA}$ |
|--------------------|-----------------------|----------------------|----------------------|------------------------------|----------------------------|
| n=2 | 22.3 (4) | 18.7 (5) | 12 (2) | 8.9 (2) | 10.4 (3) |
| n = 4 | 22.1 (2) | 20.0 (2) | 11(1) | 5.2 (2) | 6.6 (2) |
| n = 8 | 21.6 (3) | 20.1 (2) | 12(1) | 5.9 (3) | 7.6 (4) |
| n = 20 | 22.9 (2) | 19.4 (2) | 11 (*) | 5.7 (2) | 7.5 (3) |
| n = 40 | 21.8 (2) | 20.2 (2) | 12 (*) | 5.5 (2) | 7.0 (3) |

roughness obtained by AFM for the starting growth surface. Such figures are within 1% variation with respect to the electron densities obtained from room temperature phase unit cells for bulk materials. This indicates that although the deposited layers are amorphous—as measured by x-ray diffraction (not shown here)—they do not exhibit vacancies that could lead to mass density reduction, pointing out to a scenario compatible with conformal growth.

Figure 2(a) shows reflectivity profiles for samples of series A with n=2 and n=4. For the n=2 case, one cannot distinguish satellite peaks, since the total amount of stacked layers is very reduced. The profile is then dominated by the oscillations that correspond to the total thickness D. For n=4, satellites start to appear, indicating that the layers in



FIG. 2. (Color online) (a) XRR measurements for n = 2 and n = 4 ($T_g = 140 \,^{\circ}$ C) samples (dots), (b) XRR measurements for n = 20 and n = 40 ($T_g = 140 \,^{\circ}$ C) sample (dots). In both figures fits using the kinematical model (red solid line, high-q region) and the Parratt model (blue solid line, low-q region) are shown.

the stack are very similar in thickness, presenting well defined interfaces (low roughness). This is in fact observed in the behavior of the σ_{AlHf} parameter for this sample in Table I, which exhibits its lower value for the whole series.

X-ray reflectivity results for stacks of several layers – n = 20 and n = 40 – are shown in Fig. 2(b). It can be noticed that, while the XRR model nicely describe all features of n = 2, 4, 8, there are deviations from the model to the measured data at high order satellites. These deviations suggest that fluctuations in interface roughness may arise along the layer stack, leading to a worse average value of σ_{AlHf} in n = 20, 40, with respect to n = 4. Such nonhomogeneity cannot be captured by our model, since it would require the introduction of z-dependent layer thickness and roughness variables. Since XRR measurements present a phase problem - meaning one cannot clearly distinguish a system in which rougher layers come first from a system in which rougher layers are on the top of the stack – we believe a variable roughness/thickness model would introduce strong ambiguities in the data analysis. However, the sum of Al₂O₃ and HfO₂ layer thickness obtained by our model for all stacks of series A exhibit only small variations, about 2% with respect to the average value for the whole series, indicating that thickness fluctuations are unlikely to happen as stacking proceeds. This is not the case for the average interface roughness for the samples listed in Table I, where strong variations are seen. The evolution of roughness in sample series A will be further explored by comparing XRR and TEM results in the end of this section.

In order to investigate the effect of different growth temperatures on the interface roughness for Al₂O₃/HfO₂ multilayers, we have performed XRR measurements in samples with n = 8 grown at 110°C, 140°C, and 180°C. The results are shown in Fig. 3. It is clearly seen in these results that although the first satellite peak and its vicinity have the same shape for all samples, drastic changes are observed in the second satellite peak. While such feature is very well defined in the sample grown at 180°C, it vanishes at the sample grown at 110 °C. This behavior was captured by the model fitting. Similarly to the first series, the electronic density of all compounds were kept constant, with the same values used for n = 2, 4, 8, 20, 40 with $T_g = 140$ °C.

From Table II, that summarizes the results for the n = 8temperature series B, some conclusions can be drawn. High growth temperature (180 °C) produces a multilayer with reduced interface roughness, most probably due to the enhanced mobility of the precursor molecules on the surface. This allows for more uniform coverage since local energy minima for adsorption can be achieved more efficiently. In the sample grown at 110 °C the thickness of both Al₂O₃ and HfO₂ was found to be the same, which accounts for the suppression of the second satellite in the XRR curve (the Fourier transform of a perfect square wave exhibits only odd maxima). A more pronounced thickness difference is observed as the growth temperature rises until 180 °C. This is shown by the raising of the second satellite, and may imply that the growth conditions are being shifted from a thermodynamic window that favors the adsorption of the HfO₂ precursor, toward a condition that favors the reaction of trimethyl-alumi-



FIG. 3. (Color online) Measured x-ray reflectivity curves (dots) for n = 8 multilayers grown at 110 °C (bottom), 140 °C (middle), and 180 °C (top). The solid red (q < 0.15Å⁻¹) and blue (q > 0.15Å⁻¹) lines are fits using the kinematical and Parrat model, respectively. The results from these fits are shown in Table II.

num (Al₂O₃ precursor). The position of the third satellite peak, more sensitive to deviations in the Al₂O₃/HfO₂ bilayer thickness is shifted, directly evidencing that thicker bilayers are obtained at 110 °C – $d_{bilayer} = 42.7$ Å – while larger growth temperatures produce thinner bilayers: $d_{bilayer} = 41.7$ Å for 140 °C and $d_{bilayer} = 41.3$ Å for 180 °C.

For the sample series B the evaluation of Al_2O_3/HfO_2 interface roughness evidences a reduced variation with respect to sample series A. As discussed above, surface precursor mobility is enhanced at high deposition temperatures for the series. The result obtained by back heating, used during the synthesis, may be comparable to indirect heating of the reactor chamber since a long thermalization time takes place prior to the layer deposition and no relevant thermal gradient is expected in the vicinity of our thin samples (substrate thickness < 1mm). Thermally induced changes in roughness are hence expected only due to the time interval in which samples stayed inside the reactor, and will be

TABLE II. Thickness and roughness parameters extracted from XRR fits to sample series B, with $T_g = 110$, 140, and 180 °C for a fixed number of HfO₂/Al₂O₃ bilayers n = 8.

| Temperature of deposition (°C) | $d_{AI2O3} {\rm \AA}$ | $d_{HfO2} {\rm \AA}$ | $d_{SiO2} {\rm \AA}$ | $\sigma_{ m AlHf}{ m \AA}$ | $\sigma_{ m HfAr}{ m \AA}$ |
|--------------------------------|-----------------------|----------------------|----------------------|----------------------------|----------------------------|
| 110 | 21.4 (3) | 21.3 (3) | 12(1) | 5.6 (2) | 7.1 (5) |
| 140 | 21.6 (2) | 20.1 (3) | 12(1) | 6.0 (3) | 8.2 (4) |
| 180 | 22.5 (3) | 18.8 (2) | 12 (1) | 5.5 (2) | 6.8 (3) |

discussed in the following paragraphs. Finally, changes in thermal conductivity are expected for multilayer stacks with large number of periods.⁹ Such issue is of crucial interest for applications in which the interface roughness may be affected by a high local current density that can induce temperature gradients on a device. In such scenario, recrystallization processes are the main source of local defects, since the re-arrangement of atoms from the initially amorphous layers very often leads to faults, dislocations and in some cases mixing. The atom mobility necessary for this local rearrangement is achieved at \sim 500 °C for HfO₂ (Ref. 5) and ~ 1000 °C for Al₂O₃.^{7,8} Therefore, at the temperatures used during the synthesis no pronounced mixing of layers - that would indicate larger mobility - was observed (see microscopy results below). Large roughness changes would then be expected only in conditions in which the local temperature approaches the recrystallization for one of the compounds.

Although the x-ray reflectivity analysis provides unambiguous average values of roughness and thickness for the multilayer stacks studied here, it does not give a direct view of the interface morphology. Up to the present point, we were able to detect variations in the interface quality as deposition parameters changed, but local inhomogeneities such as stacking defects cannot be captured by our models. In order to search for these inhomogeneities TEM measurements were carried out. Figure 4(a) shows a TEM image with a large field of view for sample with n = 40, $T_g = 140$ °C. One can notice the strong contrast from the dark layerslarger electronic density, corresponding to HfO2-to the bright layers (Al_2O_3) in the stack. A close look at this sample can shed light into the roughness in the sample series in which n ranges from 2 to 40. The first layers, which are deposited on top of the Si/SiO₂ substrate shows flatter interfaces with respect to the upper layers. This is in agreement with the measured AFM roughness for the starting surfaces $(\sigma_{\text{substrate}} = 2 \text{ Å})$ and with the larger average roughness from the HfO₂/Al₂O₃ interfaces determined by XRR. However, this does not reflect on a better roughness for the n = 2 sample. We speculate that the annealing of the lower interfaces during the deposition of n = 40 layers (the sample growth takes about 10 hs) the bottom interfaces have enough energy and time to better adapt to the improved substrate roughness.

One sees that upper layers exhibit a larger fluctuation of interface position with respect to the bottom layers. From the TEM results one also observes that in-plane variations of the layer interface position are reduced at the initial (bottom) layers as compared to the uppermost layers. The larger periodicity in interface fluctuation along the in-plane direction for the upper layers may have a crucial influence in the decrease of average interface roughness observed in XRR for the n = 40 sample, since locally these interfaces will be less abrupt than the interfaces for the middle of the stack (corresponding to smaller n values). The inset of Fig. 4 shows a Fourier transform of the multilayer image, exhibiting well defined subsidiary satellites which correspond to a periodic system in the growth direction with a periodicity of 42 Å, matching the bilayer periodicity observed by x-rays.

Figure 4(b) shows a zoom in the region limited by the dashed box in Fig. 4(a). In this region one observes a stack-



FIG. 4. (Color online) (a) TEM image of a cross-section of the n = 40 multilayer. The inset shows a Fourier transform for panel (a). The detail of a stacking defect marked by a red dashed box in (a) is shown in (b). (c) Geometrical phase image using a mask comprising the first fast Fourier transform satellite periodicity and the range inside the green dotted square shown in (a) as reference. The spatial resolution was set to 13nm. (d) Distortion map on the bilayer spacing, obtained from (c). (e) Fringes rotation map obtained from (c).

ing defect of the layers. In this stacking defect, the sequence of Al_2O_3/HfO_2 layers along four periods does not match locally the expected multilayer profile. This kind of defect was observed at different positions of the n = 40 sample, which was studied by TEM. Although the density of these defects is not representative to cause changes in the XRR measurements, it can affect the performance of devices in which a reduced number of very thin alternated layers are used to provide hybrid characteristics such as good electric insulation and mechanical robustness. Therefore, the deposition of several layers can be used to avoid this kind of stacking fault in systems that require high reliability of the oxide material properties.

Besides the local stacking defects shown in Fig. 4(b), fluctuations in periodicity and local layer alignment with respect to a reference can be extracted by geometric phase analysis (GPA) of TEM images. This method has been successfully used to study strain and defects in crystalline lattices^{17,18} but can also be applied to understand variation in nm-modulated systems like ours. Figure 4(c) shows the phase analysis for the central subset of sample range shown in Fig. 4(a), using a mask corresponding to the frequency of the first satellite of the fast Fourier transform image and the position marked by a solid line square (green) in Fig. 4(a) as reference. Such reference region is obtained from the analyzed image and was carefully chosen for having a minimum geometrical phase change, i.e., reduced variations in periodicity and rotation. In this case the phase mapping resolution in real space is set to 13 nm, and from the different tones on Fig. 4(c) one can observe smooth changes in the multilayer periodicity that take place in the same length scale of the chosen phase window. Therefore, besides local stacking defects and layer roughness, fluctuations on the bilayer periodicity, which involves the thickness, are present along the interface direction.

Based on the phase mapping, it is possible to obtain a fringe distortion (or deformation) and rotation maps with respect to the selected reference region, which are shown in Fig. 4(d) and 4(e), respectively, for the same sample region. In such fringe distortion map the fluctuations in the fringes periodicity along both the interface and the stacking directions are clearly observed. The distortion map reveals the presence of two defects, marked by arrows. One of these defects could be easily found in the original image, as shown in Figs. 4(a)-(b), while the other just became clear under GPA analysis. The slope of layers also changes within the sample, as captured by fringes rotation analysis of Fig. 4(e). Such changes may arise from the conformal covering obtained during ALD growth. Punctual changes in thickness are continuously buried by new conformally deposited layer as the stacking proceeds. The effect of such procedure is the generation of layers in which the local alignment to the substrate interface reference line is distorted. Angular tilts of $\pm 10^{\circ}$ are observed in Fig. 4(e). Such tilting of layers is already qualitatively observed in the angular broadening of the



FIG. 5. (Color online) (a) TEM image a different region of the n = 40 multilayer. (b) Bilayer spacing distortion, clearly showing a region with periodicity fluctuation with respect to the rest of the multilayer image.

first satellite of the Fourier transform inset of Fig. 4(a) with respect to the main periodicity direction, which indicates a variation of the slope of the interfaces with respect to the average slope.

The ability of the GPA method to reveal stacking defects in this multilayer system can be better appreciated in Fig. 5. From the stacking sequence of Fig. 5(a) one cannot directly tell whether a thickness fluctuation is effectively taking place in any region of the image. Finding such fluctuation becomes even harder due to the roughness of the layers. However, by performing GPA analysis on Fig. 5(a), a large region where the bilayer thickness becomes different from the reference state is observed.

V. CONCLUSION

Atomic layer deposition of oxide materials has become a standard technique for deposition of thin oxide films for electronic device research and industrial processes. Despite of its great achievements of coverage uniformity and scaleup processes, the complexity of ALD-based devices is continuously increasing. Many of the foreseeing applications will have constraints that go beyond the simple requirements of homogeneous coverage and conformality. As the layer thicknesses reduce to explore surface/interface effects the relevance of parameters such as roughness and thickness fluctuation becomes crucial.

As discussed here, despite of presenting constant thickness, ALD short-period multilayers exhibit global and local changes in roughness and layer interface alignment, among other parameters. For instance, the average roughness of the n = 2 bilayer sample studied in this work was much larger than the values obtained for lager stacking sequences (A series), indicating that the stacking process and longer annealing/growth time plays a role on interface smoothening. The n = 8 bilayers temperature series (B series) explored in this work also points out that although ALD depositions take place at considerably low temperatures, as compared to MBE/CVD/sputtering processes, the interface quality can be slightly improved under fine tuning of deposition parameters. It has also been shown here that for conditions in which the roughness and thickness fluctuations are close to the layer thickness values, the exact Parratt model for reflectivity does not describe correctly the average behavior of the system. Such average evaluation is crucial since the possibility of using a z-dependent roughness profile introduces too many variables to XRR analysis, which may lead to unreliable results.

From the TEM observation and GPA analysis of multilayer local configuration one can infer that devices based on local properties may respond differently from those in which the behavior of the ensemble is dominant. For such nanotech-devices where short-period layers would be used, stacking defects and local thickness/layer alignment distribution can strongly affect properties of interest. It is crucial, therefore, to investigate both the evolution of stacking effects on layer quality, as well as substrate surface-dependent parameters that may propagate inhomogenities on multilayer growth. M. de Pauli acknowledges FAPESP project 2009/11875-2 and the CNPq/LNLS summer internship program. A. Malachias acknowledges FAPESP project 2009/09027-3. Y.F. Mei acknowledges the support from the National Natural Science Foundations of China.

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