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Effect of aluminum nitride buffer layer deposited by molecular beam epitaxy on the growth of aluminum nitride thin films deposited by DC magnetron sputtering technique

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Abstract: This paper reports the effect of silicon substrate orientation and aluminum nitride buffer layer deposited by molecular beam epitaxy on the growth of aluminum nitride thin films deposited by DC magnetron sputtering technique at low temperature. The structural analysis has revealed a strong (0001) fiber texture for both substrates Si (100) and (111) and a hetero-epitaxial growth on few nanometers AlN buffer layer grown by MBE on Si (111) substrate. SEM images and XRD characterization have shown an enhancement in AlN crystallinity thanks to AlN (MBE) buffer layer. Raman spectroscopy indicated that the AlN film was relaxed when it deposited on Si (111), in compression on Si (100) and under tension on AlN buffer layer grown by MBE/Si (111) substrates, respectively. The interface between Si (111) and AlN grown by MBE is abrupt and well defined; contrary to the interface between AlN deposited using PVD and AlN grown by MBE. Nevertheless, AlN hetero-epitaxial growth was obtained at low temperature (<250°C).

Keywords: Hexagonal AlN, thin films, Direct current magnetron sputtering, Texture, fiber, heteroepitaxial growth.

1. Introduction

Aluminum nitride (AlN) thin films can be a promising candidate in optical, mechanical, and electronic applications. It can serve as a semiconductor when doped [1] and also as a passivation layer for semiconductors [2]. Besides, AlN thin films are integrated in surface acoustic wave (SAW) devices, where they insure high frequency ranges, a large electromechanical coupling factor ($\boldsymbol{K_s^2}$) and temperature stability of the corresponding device. Nevertheless, the quality of these integrated films has a strong impact on the performance of the SAW devices [3-4]. The film's properties depend not only on the crystal structure of AlN but also, on its preferential orientation [5], c-axis in our case. The films can be composed of multiple crystal orientations scored (002), (100) etc. AlN thin films are grown, using physical vapor deposition (PVD), on several substrates such as silicon (Si) [6], sapphire [7], and indium phosphide (InP) [8]. However, depositing AlN layers on Si simplifies the process and the device structure for a low cost [9].

It is well known that GaN is one of the most important semiconductors. It is used in blue and ultraviolet light emitting devices as well as high-temperature and high-frequency, high-power electronic devices owing to their excellent properties [10]. Electronic and optoelectronic devices based on GaN deposited on Si (100) and Si (111) substrates are thus highly attractive. GaN films have the potential to be integrated with well-developed Si microelectronic circuits [11]. On the other hand, it is very difficult to

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realize high-quality epitaxial GaN layers on Si. One method for this is to cover the Si substrate by using AlN as a buffer layer, which serves as a substrate for further GaN growth. The AlN has a wide band gap (6.2 eV) III–V compound with high value of thermal conductivity, chemical and thermal stability, refractive index, and breakdown dielectric strength. With these properties, the thermodynamically stable Wurtzite AlN films are not only a good buffer layer materials for GaN growth but also a promising material for applications in other microelectronic and optoelectronic devices [12-14].

In this study, the AlN thin films were deposited using a reactive direct current magnetron sputtering technique (DCMS). AlN films were grown on Si (100), Si (111) substrates and 1 nm AlN grown by MBE on Si (111) as interlayer. The effect of substrate type on the microstructure and quality of deposited films was studied. It is worth to note that the AlN interlayer grown by MBE has a very good quality with a rocking curve RC-FWHM less than 0.3° for 150 nm thick and surface roughness of 1 Å [15]. This interlayer has high electrical resistivity and good thermal conductivity.

The aim of this work is to show that is possible to obtain a hetero-epitaxial growth of AlN films deposited by DCMS method at low temperature on Si (111) by using a few nm AlN buffer layer grown by MBE. The low temperature is very important in order to reduce the thermal expansion problem existing between AlN and GaN. This work is dedicated to the growth of aluminum nitride thin films by PVD techniques at low temperature (<250°C) and obtain thicknesses up to micron. Such thick and low temperature AlN films will be used in order to ensure thermal management of power electronic devices.

2. Materials and Methods

AlN films were deposited using a DCMS technique. An 8″ target was used consisting of 99.99 pure aluminum (Al) water-cooled magnetron cathode. The system was pumped to a base pressure of 10^{-8} Torr with a turbo molecular pump before introducing argon (Ar) and nitrogen (N_2) gases. The sputtering system used to deposit AlN films is a Pinnacle Plus+ 5kW® DC power supply, with a power of 1800 W. The target was cleaned before deposition using Ar gas discharge followed by a pre-sputtering step using the same conditions as the subsequent film deposition with a shutter shielding the sample in order to remove surface oxidation of the target. The sputtering chamber was evacuated to 2.2 mTorr which is fixed as base pressure. The reactive N_2 gas was fixed at 55%.The structural characterization of AlN films was performed by X-ray diffraction (XRD) using a PANanalytical Empyrean® X-ray diffractometer, with Cu Kα radiation (λ = 0.154 nm), voltage and current (40 kV - 40 mA), respectively. For more details check [16]. In addition, Raman spectroscopy was used in order to obtain the quality of the deposited films and calculate the residual stress which has been developed after deposition. The reflection spectra of the respective molecular structure for the prepared AlN films are identified, studied and presented using Raman spectrometer (Horiba Jobin-Yvon lab-RamT64000®) with the green laser of 514 nm excitation at room temperature. Moreover, scanning electron microscopy (SEM) and the High-Resolution Transmission Electron microscopy (HRTEM) were used in order to thoroughly study the films microstructure, the morphology and the interfaces between the layers. The HRTEM is used only in the case of the AlN deposited on AlN MBE/Si (111).

3. Results and discussion

The XRD patterns of the θ-2θ (Bragg-Brentano Geometry) and the θ-θ scans of AlN/Si (100), AlN/Si (111) and AlN/AlN MBE (1 nm)/Si (111) are shown in Figure 1 (a) and (b), respectively. The highest intensity of the (0002) reflection at $2\theta=36.15°$ indicates an oriented growth along the *c-axis* perpendicular to substrate in both AlN films depos-

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ited on AlN buffer layer and Si (111). The measure of peak width, the full width at half maximum of the X-ray rocking curve (RC-FWHM) for a determining the quality of the film (Gaussian curve). It can be observed that the FWHM values decreases from 3.2° to 2.3°respectively. This result gives an indication that the quality of the AlN films has been improved depending on the Si orientation. On the other hand, the diffraction peak intensity of the (0002) AlN is increased and the RC-FWHM of the AlN film decreased from 2.3° to 1.2° using the AlN (MBE) interlayer, this can indicate that the use of AlN buffer layer enhance the crystalline quality of AlN film and facilitate its deposition. In addition, there is a shift in the peak position of AlN (0002) orientation between the film deposited directly on Si (111), where 2θ = 36.09°, and that deposited on AlN MBE (1nm)/Si (111), where 2θ = 36.15°. This shift is due to the difference in the residual stress developed inside the AlN films.

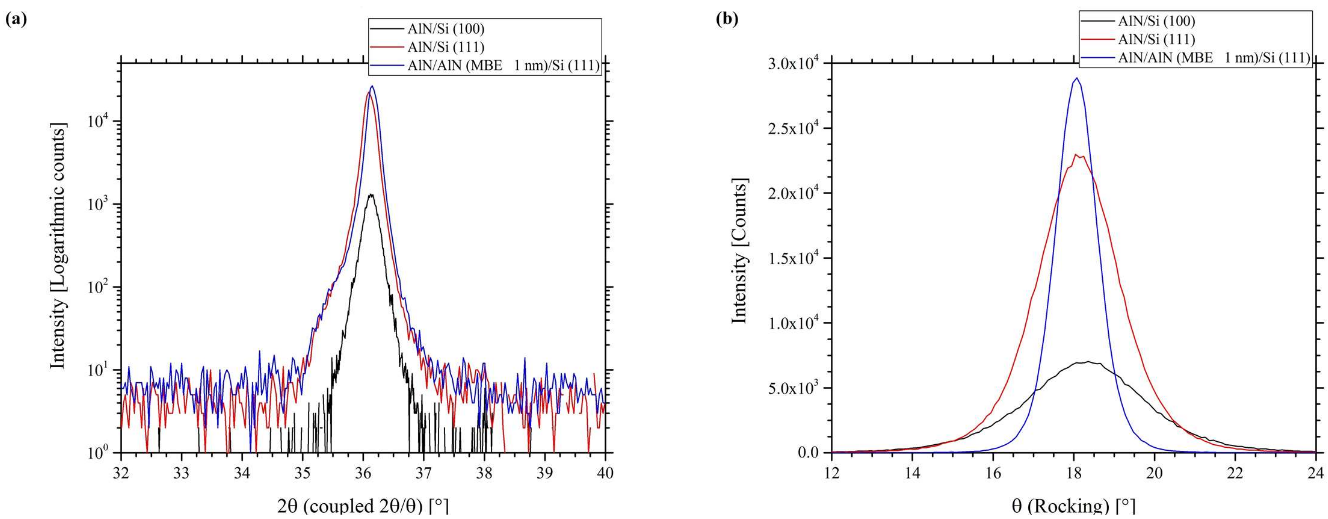

Figure 1. Diffractogram in the (a) θ-2θ mode and in the (b) θ-θ mode showing the AlN peak of (0002) orientation for the samples.

To understand this change, XRD pole figures (0001), $\{10\bar{1}1\}$, $\{10\bar{1}2\}$ and $\{10\bar{1}3\}$ of h-AlN were obtained, but only the pole figure $\{10\bar{1}1\}$ and $\{10\bar{1}2\}$ are used in this study. We characterized, in reflection mode, the incomplete pole figures for χ= 0 to 75° (tilt angle); beyond, the defocus problem becomes too important. The step size is 2.5° for both angles χ and φ (azimutal angle). The $\{10\bar{1}1\}$ pole figures of AlN/Si (100), AlN/Si (111) and AlN/AlN MBE/Si (111) films are shown in Figure 2a, b and c respectively.

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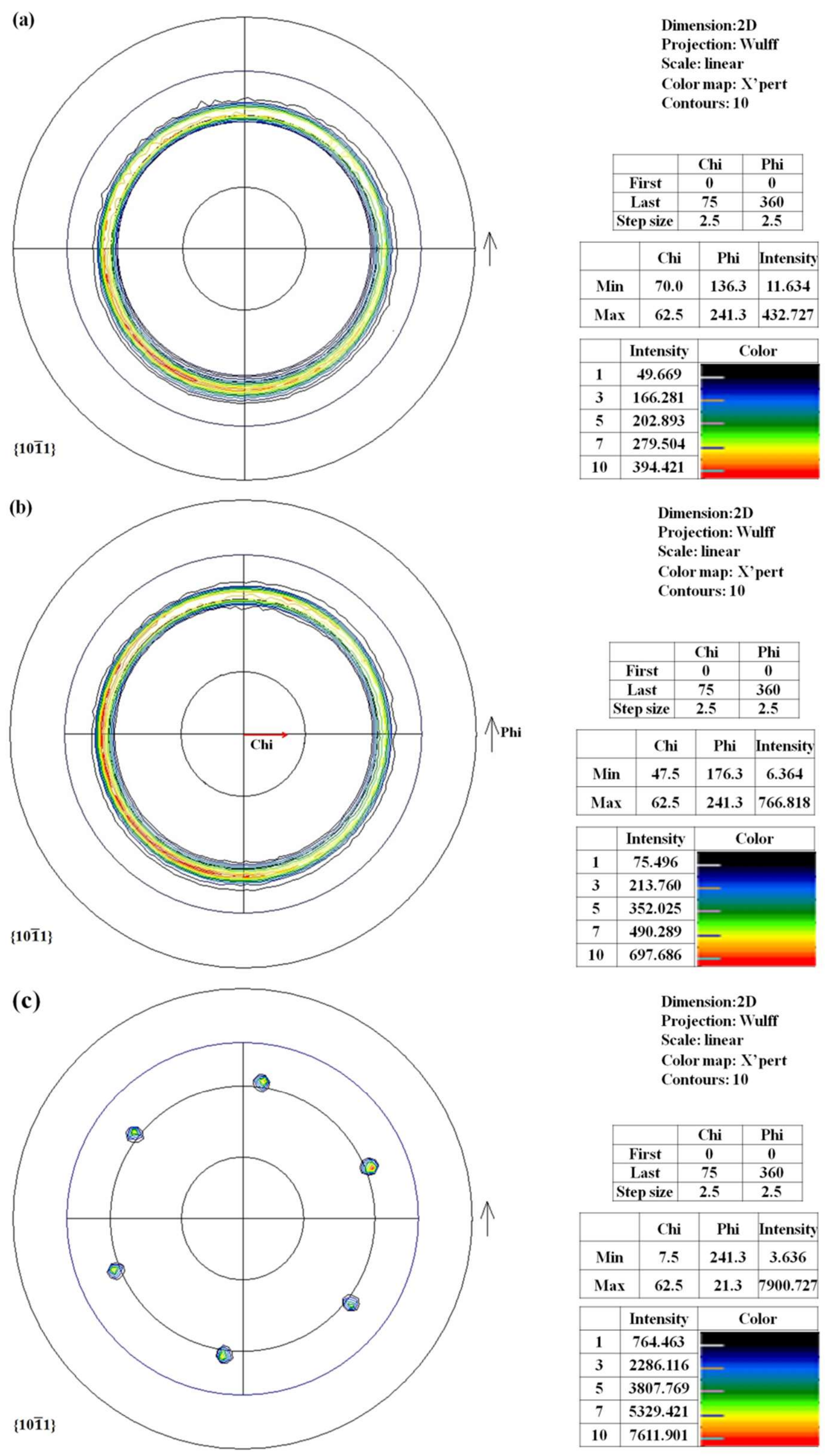

Figure 2: {10$\bar{1}$1} pole figures (2θ=37.89°) of (a) hexagonal AlN/Si (100) (b) hexagonal AlN/Si (111) and (c) of h-AlN/AlN MBE (1 nm)/Si (111).

For both AlN/Si (100) and AlN/Si (111) samples, the {10$\bar{1}$1} pole figures intensity maxima are distributed along a ring located at χ = 62.5°, demonstrating the presence of a polycrystalline film with (0001) strong fiber-texture, which the grains of the thin film are

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composed of one family of parallel planes at the substrate's surface and having an axis of rotation around the normal of these planes. Moreover, the ring at $\chi = 62.5^{\circ}$ caused by the $\{10\bar{1}1\}$ facets implies that no preferred in-plane orientation is formed for both cases. On the other hand, the width of the ring is related to the dispersion of (0001) orientation planes. However, in the case of AlN deposited on AlN (MBE) /Si (111), the $\{101\bar{1}\}$ pole figure shows a maximum intensity with a six-fold symmetry at χ=62.5°. It is good to mention that for a 1 nm thick AlN buffer layer a hetero-epitaxial growth of AlN deposited by DCMS method was obtained. This indicates that the AlN thin film deposited on AlN /Si (111) has not any more a fiber-texture, but has grown epitaxially on the AlN /Si (111) substrate. It can be noticed that the addition of an AlN buffer layer between the deposited AlN PVD and Si (111) substrate allows a hetero-epitaxial growth of the AlN thin films. This can be explained by the lattice mismatch between AlN and Si (100), which is much higher than that with Si (111); thus, the AlN film growth is easier on Si (111). It can be seen that almost 0% mismatch exist between AlN and AlN buffer layer /Si (111), 19% between AlN and Si (111) substrate and 43% in the case of AlN film and Si (100) substrate [16-18].

The morphological and cross sectional characterization of AlN films synthesized at different substrates Si (100), Si (111) and AlN buffer layer/Si (111) were realized using SEM analysis. The cross-section SEM images of deposited films displayed in Fig 3a, b and c. Fig 3a showed an abrupt interface between Si (100) and AlN films deposited by PVD, the AlN layers exhibited an irregular columnar growth with cracks. Fig 3b presented also an abrupt interface between Si (111) and AlN films but it can be seen that there is an inter diffusion between Si substrate and sputtered AlN films with dense columnar structure. However, Fig 3c exhibited the AlN film deposited on AlN buffer layer/Si (111) substrate. This later has an almost regular columnar structure with clear abrupt interface between AlN buffer layer and AlN PVD without defects and cracks, also good film crystallinity. It can be seen that the AlN film deposited on AlN buffer layer/Si (111) has the best columnar structure compared to those deposited on both Si (100) and Si (111) substrates. This confirms the results of XRD analyses shown previously.

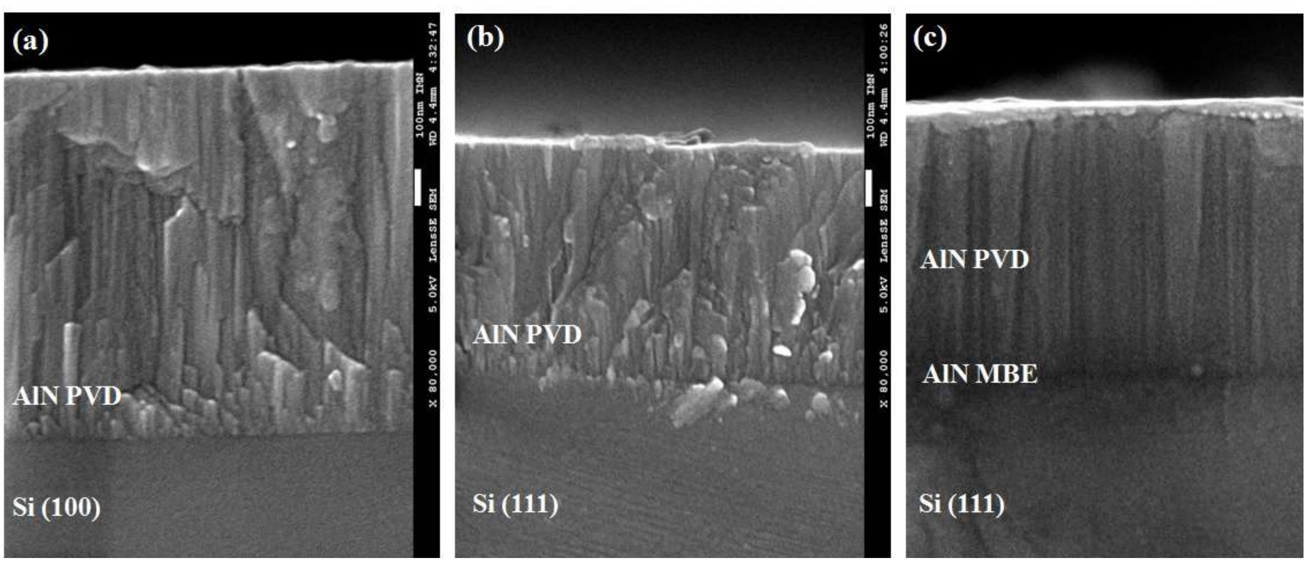

Figure 3. Cross section SEM image for (a) AlN/Si (100), (b) AlN/Si (111) and (c) AlN/AlN MBE/Si (111).

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The samples were also examined by visible Raman spectroscopy at a wavelength of 514 nm and a full laser power. Raman results provides additional information about the AlN film's quality via the calculation of the High bands (E_2^H) of the AlN films. Moreover, the (E_2^H) mode position gives an indication about the residual stress within the film. Figure 4a shows the Raman spectra with AlN bands A_1 (TO) and E_2^H for the three samples. One can observe a shift of few nanometers of the E_2^Hmode. Moreover, the residual stress developed inside the deposited films was calculated using radius curvature method thanks to Profilometer tests and such data were coupled with Raman shifts. The variation of the stress as function of the (E_2^H) band shift is represented in Figure 4b.

It can be seen from Figure 4 that the AlN film deposited on Si (111) substrate is almost relaxed (Stress = 0.03 GPa). Nevertheless, for AlN deposited on Si (100), the stress is equal to -0.37 GPa meaning that the film is in compression. The strain difference in the AlN films deposited on the two types of the substrates can be attributed to the extent of lattice mismatch [17, 19]. On the other hand, AlN films deposited on AlN buffer layer/Si (111) have shown a stress of 3.23 GPa. This result indicates that the film is under tension.

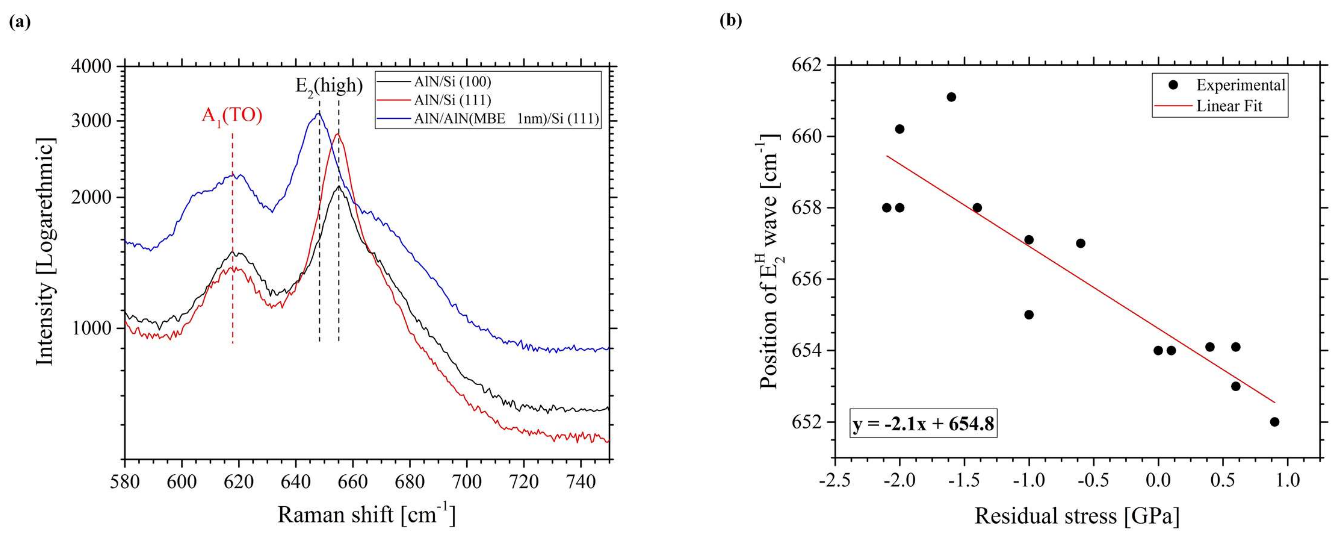

Figure 4. (a) Raman spectra of AlN thin films and (b) Position of the Raman band relative to the AlN (0002) orientation as a function of the residual stress calculated by the method of radius of curvature on Si substrate.

Fig. 5 shows TEM cross section analysis of thick AlN film deposited on few nanometers AlN epitaxial film on Si (111) using DCMS technique. Figure 5 (b) and (c) show a cross section TEM image at low magnification of the AlN layers and Si interface. The contrast differences are observed on 3 nm from the Si substrate. This difference on the contrast can be representative to the difference between the compositions related to the surface oxidation. It is very important to notice that the AlN thin films and AlN buffer layer are deposited in two different reactors. We have chosen that the surface of the buffer layer was not cleaned to avoid its amorphisation (oxidation). From the TEM images, it appears that the upper AlN exhibits a columnar structure, a very good continuity is observed between both films. The sputtered film is also continuous layer of 3 nm thick. This layer corresponds to the 1 nm AlN MBE layer already measured by RHEED technique [20]. Thus, it is obvious that there is an underestimation of the thickness. Moreover, this image confirms that the interface between AlN buffer layer and Si is abrupt. Also, a dense structure is highlighted for the sputtered AlN films followed by column like morphology.

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On the other hand, the interface between the AlN (MBE) buffer layer and AlN deposited by PVD is less abrupt. This is probably due to the presence of defects at the interface. These defects are probably caused by contamination of the buffer layer before AlN sputtering as the two AlN films were deposited in two different reactors. Despite such imperfect interface AlN hetero-epitaxial growth has been reached at low temperature by PVD method.

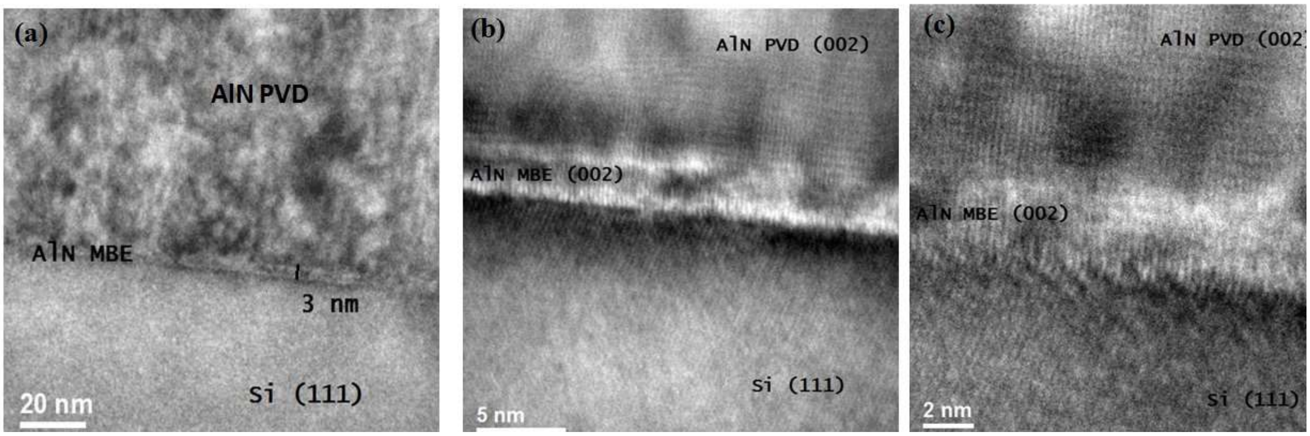

Figure 5. Cross-section HTTEM images at different magnification for the interface between the AlN PVD and the AlN MBE.

5. Conclusions

AlN films were grown on Si (111), Si (100) and MBE grown AlN buffer layer on Si (111) substrates with reactive direct magnetron sputtering (DCMS) process at low temperature. The AlN film structures, obtained for all substrates were investigated by XRD, SEM, Raman and TEM analysis and compared. The crystallinity of AlN layers was improved using 3 nm AlN buffer layer. This layer was found to promote the formation of c-axis oriented AlN layers and suppress the formation of disoriented crystallites on the non-c-axis planes, resulting in the improved crystallinity. Nevertheless, it is thought that only the surface state (oxidation, nitriding, and restructuring) remains an obstacle to a hetero-epitaxial growth of hexagonal AlN directly on Si substrates. The FWHMs of the (0002) X-ray rocking curves were decreased from 3.2° to 1.2° using AlN buffer layer. The texture analysis has revealed a strong (0001) fiber texture for both substrates Si (100) and (111) and a hetero-epitaxial growth on 1 nm AlN buffer layer grown by MBE on Si (111) substrate. It appears that the AlN is respectively relaxed, under compression and under tension when it deposited on Si (111), Si (100) and AlN buffer layer. HRTEM confirmed that the interface between AlN grown by MBE and Si (111) is abrupt. On the other hand, it indicated an unclear interface between the AlN grown by MBE and the AlN deposited by PVD, probably due to the presence of defects. Nevertheless, despite such imperfect interface AlN hetero-epitaxial growth has been reached at low temperature by PVD method.

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