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Study of high quality AlN layers grown on Si(111) substrates by plasma-assisted molecular beam epitaxy

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Abstract

High quality AlN layers with full widths at half maximum values of 10 arcmin and average surface roughness (rms) of 48Å were grown by molecular beam epitaxy on Si(111) substrates. A systematic study and optimization of the growth conditions was performed in order to use these AlN layers as buffers in the growth of GaN films. Atomic force microscopy (AFM) and X-ray diffraction (XRD) techniques were employed to determine the surface and structural quality of the layers. Best AlN films were obtained at high substrate temperatures (T_{subs}>900°C) and III/V ratios close to stoichiometry. Growth conditions with III/V ratios beyond stoichiometry (Al-rich) did not further improve the crystal quality. In these cases a higher substrate temperature is needed to prevent condensation of Al on the surface. GaN films with full width at half maximum of 10 arcmin and improved optical properties were grown on top of optimized AlN buffer layers.

1. Introduction

Growth of group III nitrides has received a great deal of attention during the last few years because of their potential as materials for optoelectronic devices in the blue to ultraviolet spectral range. Room-temperature continuous-wave operation of InGaN multi-quantum-well laser diodes have already been reported on sapphire substrates by metal organic chemical vapor deposition (MOCVD) techniques [1]. However, silicon-based nitride epitaxy offers the potential for cointegration of wide band-gap optoelectronics devices with large scale circuits employing silicon-based technology. A common approach in the growth of GaN films is the use of buffer layers to improve the quality of the material [2] [3]. It is important to obtain very smooth and low defect density buffers on which to grow high quality GaN [4].

In this work we present a systematic study of the growth conditions of AlN on Si(111) by plasma assisted molecular beam epitaxy (MBE). Atomic force microscopy (AFM) and X-ray diffraction (XRD) techniques enable the assesment of the material quality. Optimization of the growth conditions leads to high quality AlN layers which are then used as buffer layers in the growth of GaN films.

2. Experimental

AlN films were grown by MBE on Si(111) substrates which had been previously cleaned following a modified Shiraki procedure. Substrates were heated at 930°C for 20 minutes in the growth chamber until a 1x1 reconstruction appeared. When lowering the substrate temperature to 780°C, a 7x7 reconstruction with prominent Kikuchi lines appeared, indicating a high quality Si(111) surface. Active nitrogen was supplied via a cryogenically cooled rf plasma source (Oxford CARS25). A photodiode is employed as an optical emission detector (OED) which generates a voltage (OED voltage) that is used as an indicative of the amount of active nitrogen in the plasma. AlN layers, grown from 780°C to 920°C under a N_2 flow of 1 sccm (growth pressure of 1.5×10^{-5} torr), had thicknesses between 0.04 and 1µm. Before starting the AlN growth, a few monolayers of Al are deposited on the substrate to avoid the formation of Si_3N_4. Once the plasma is ignited, both the Al and N shutters are opened simultaneously. A 1x1 wurtzite reconstruction is observed with elongated spots that change to a streaky pattern within 2 minutes. Under specific growth conditions this pattern gives rise to a 2x2 surface reconstruction after typically 100nm of growth. More details on the growth are given elsewhere [5].

3. Results and discussion

Figure 1 shows the dependence of AlN growth rate versus Al flux (beam equivalent pressure, BEP) for three different amounts of active nitrogen (equivalent OED voltages of 0.33V, 0.45V and 0.57V respectively). The growth rate scales with Al flux until it reaches a saturation point. Increasing the Al flux beyond that point does not increase significantly the growth rate and Al droplets start to condense on the surface. We will define that saturation point (ie. III/V ratio) as the *stoichiometry* point for a given OED value. Increases in the substrate temperature between 780°C and 920°C had almost no effect on the growth rate, but surface morphology was improved and the RHEED pattern became more streaky.

Structural characterization was obtained with high resolution XRD measurements. Figure 2 shows XRD data from AlN layers grown with different III/V ratios by increasing the Al flux at a constant OED value: from a N-rich condition (Figure 2d) to Al-rich (Figure 2a) with a close-to-stoichiometry point (Figure 2b) in between. A best value of full width at half maximum (FWHM) of 10 arcmin is observed for layers grown at/or above stoichiometry. For Al fluxes well above the stochiometry point, Al condensation might take place on the surface if the substrate temperature is not high enough. From the position of the AlN (0002) reflection a lattice constant c of 4.977 ± 0.004 Å is determined, very close to the AlN bulk constant. Therefore, all the AlN layers reach full relaxation with thicknesses as thin as 400Å.

In order to promote a two-dimensional growth of GaN, a smooth and defect free starting surface is desired. Surface analysis of the AlN layers was performed using atomic force microscopy (AFM). Figure 3 shows AFM scans of the AlN layers studied in the XRD section with decreasing Al flux. Samples shown in Figure 3b-d were grown at 850°C and sample in Figure 3a at 910°C to prevent condesation of Al on the surface. It can be seen that the rough surface of the N-rich sample, Figure 3d, becomes smoother as the Al-flux increases, reaching a best value of average roughness (rms) of 76Å in Figure 3a for this sequence. A best value of 48Å was obtained in a different series of samples.

Optimized AlN layers, in terms of structural and surface morphology, on nominally on-axis Si(111) substrates were achieved by using III/V ratios close to stoichiometry and substrate temperature around 900°C. Similar growth conditions were used on miscut Si(111) (2° towards $(2\bar{1}\bar{1})$) substrates and the result was higher FWHM values (28 arcmin) and larger average roughness (270Å), probably due to the generation of double position boundaries and/or inversion domain layers, as it was found in the AlN grown on SiC and sapphire substrates [6] [7].

In order to determine the influence of these AlN buffer layers on the quality of the GaN film, similar GaN films were grown on top of AlN buffer layers which had been grown with different III/V ratios. Figure 4a shows the XRD curve of a GaN film grown with an optimized AlN buffer layer where 0002 reflections from both the GaN and AlN layers can be appreciated. The best value for the FWHM of the GaN layer is 10 arcmin. This result is comparable to values obtained by MBE on sapphire and SiC substrates [8]. Growth of GaN directly on Si(111) substrates, with no AlN buffer layer, produced films with a high level of polycrystallinity and mosaicity (FWHM>70 arcmin).

The thickness of the AlN buffer layer is another parameter that might influence the quality of the GaN film. It has been mentioned that the RHEED pattern from the AlN exhibited a 2x2 surface reconstruction after typically 100nm of growth. Beyond that thickness the RHEED started to degrade and became spotty. Growth of GaN should start once a surface reconstruction is observed, since this indicates a two-dimensional growth with flat surfaces. Figure 5 shows XRD profiles of GaN layers grown on different thicknesses of AlN. The buffer layers used here are not fully optimized (best value of 14 arcmin for the GaN layer) but they serve as an example to verify that the quality of the GaN film improves as the thickness of the AlN increases up to 100nm. Beyond this thickness we do not expect any significant improvement due to the degradation of the RHEED pattern from the AlN layer, and in fact GaN samples grown on 500nm AlN layers show up FWHM of 50 arcmin. Finally, optical characterization of the GaN films was performed. Figure 6 shows the photoluminescence spectra of a GaN layer grown on a) an optimized and b) a non-optimized AlN buffer layer. Some improvement is clearly observed in the intensity and width of the optical emission.

In summary, high quality AlN layers were grown on Si(111) substrates by optimizing the growth conditions. III/V ratios close to stoichiometry and high substrate temperature (around 910°C) lead to a best value of FWHM of 10 arcmin. and surface roughness of 48Å (rms). These AlN layers play an important role as buffer layers, strongly influencing the quality of the GaN film. GaN layers with best value of 10 arcmin. were obtained, when optimized AlN buffers were used, indicating that Si(111) is a viable substrate for epitaxy of the wide band-gap nitrides.

Acknowledgments

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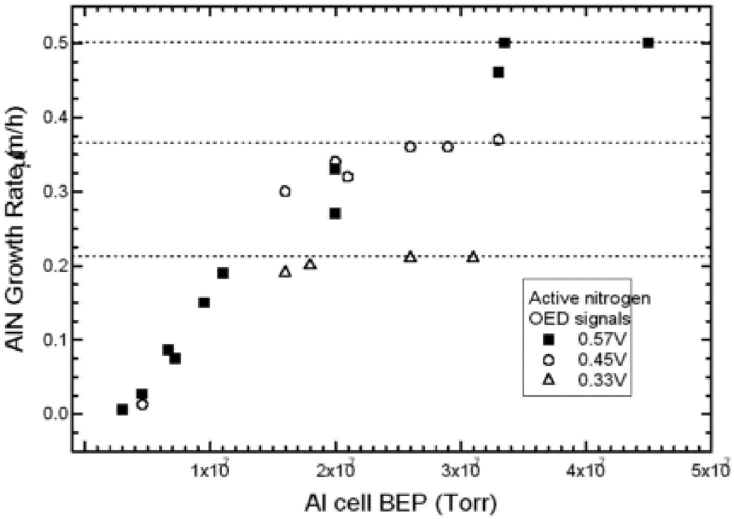

Figure 1. AlN growth rate as a function of the Al flux (Beam Equivalent Pressure) for three different amounts of active nitrogen. Substrate temperature varied from 780°C to 920°C.

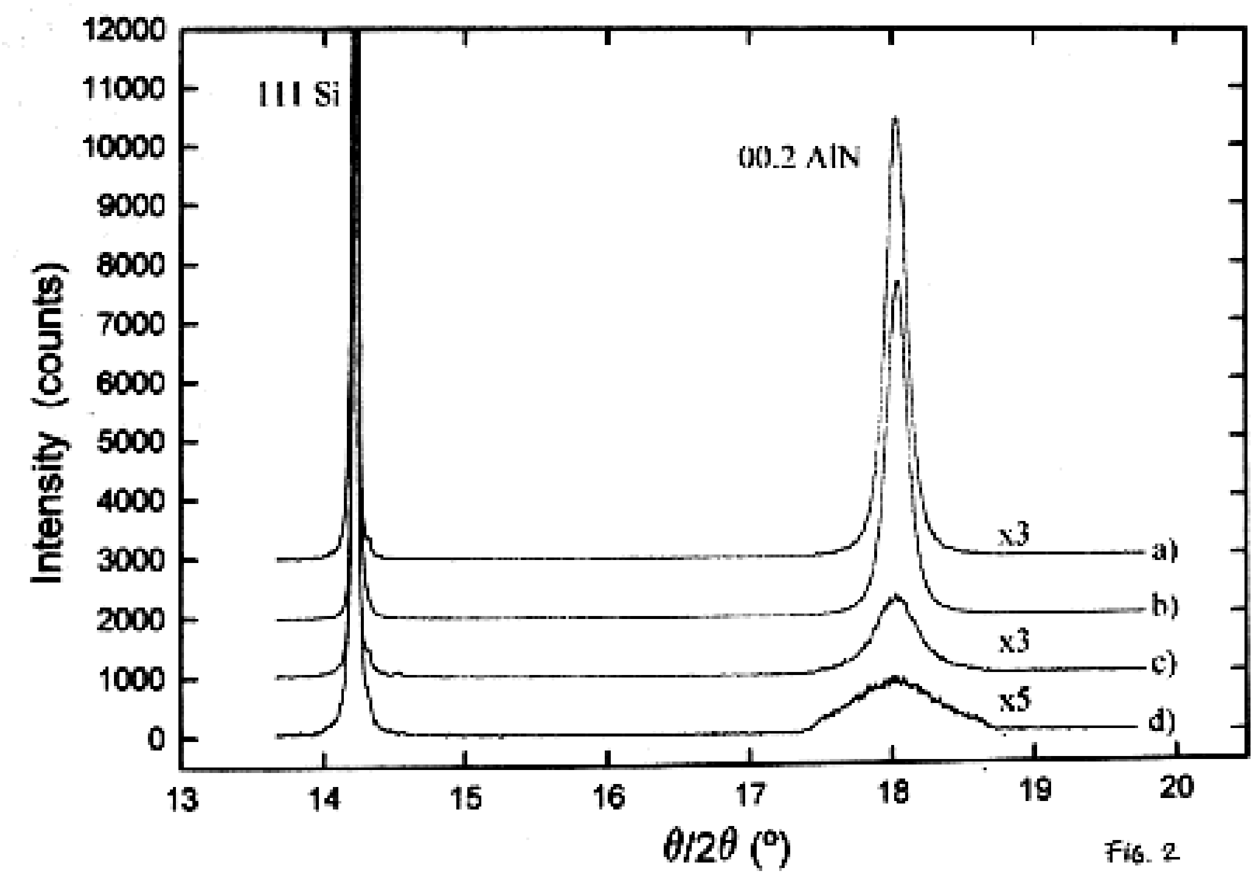

Figure 2. XRD profiles from AlN layers grown at decreasing Al flux for a given amount of active nitrogen (OED = 0.57V). a) Φ_{Al} = 4.5x10^{-7} torr (BEP), FWHM= 10 arcmin , b) Φ_{Al} = 3.3x10^{-7} torr (BEP), FWHM= 10 arcmin, c) Φ_{Al} = 2.0x10^{-7} torr (BEP), FWHM= 20 arcmin and d) Φ_{Al} = 9.7x10^{-8} torr (BEP), FWHM= 36 arcmin

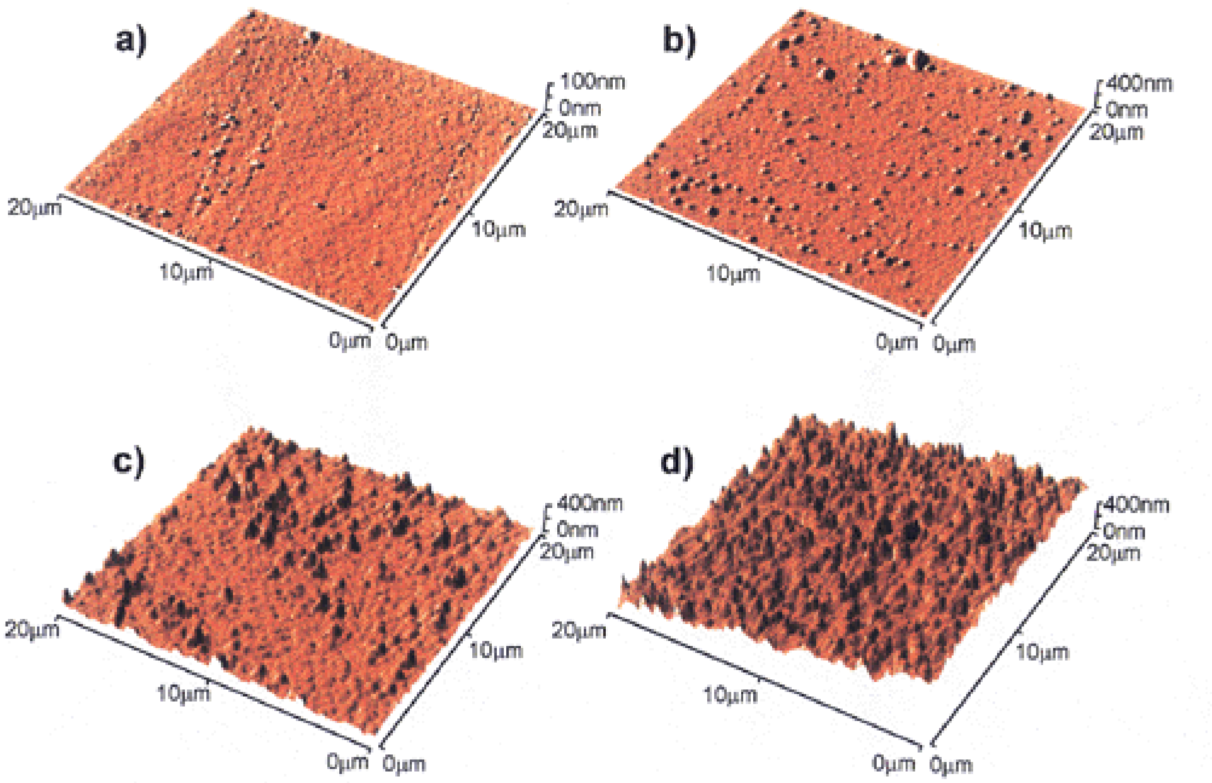

Figure 3. Surface roughness evolution, measured by AFM, of AlN layers with decreasing Al flux (same sequence of samples as in figure 2). Average roughness (rms) was: a) 7.6nm, b) 8.5nm, c) 44nm and d) 79nm.

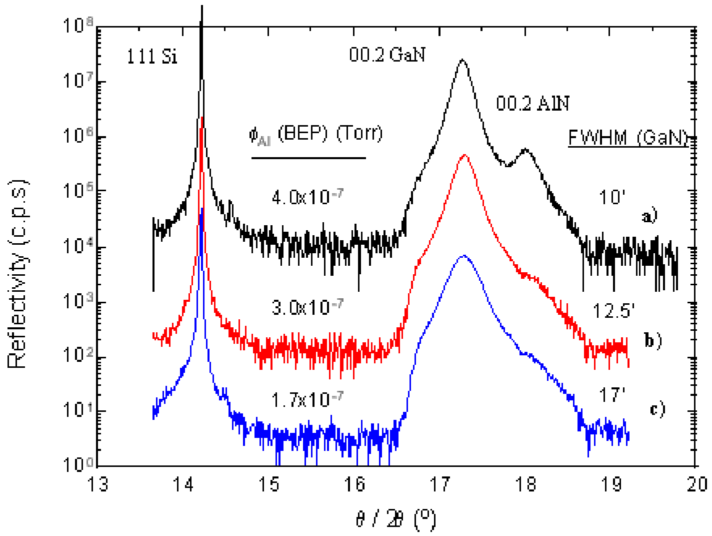

Figure 4. XRD profiles from similar GaN layers grown on different AlN buffer layers: a) Φ_{Al} = $4.0x10^{-7}$ torr (BEP), FWHM= 10 arcmin, b) Φ_{Al} = $3.0x10^{-7}$ torr (BEP), FWHM= 12.5 arcmin and c) Φ_{Al} = $1.7x10^{-7}$ torr (BEP), FWHM= 17 arcmin.

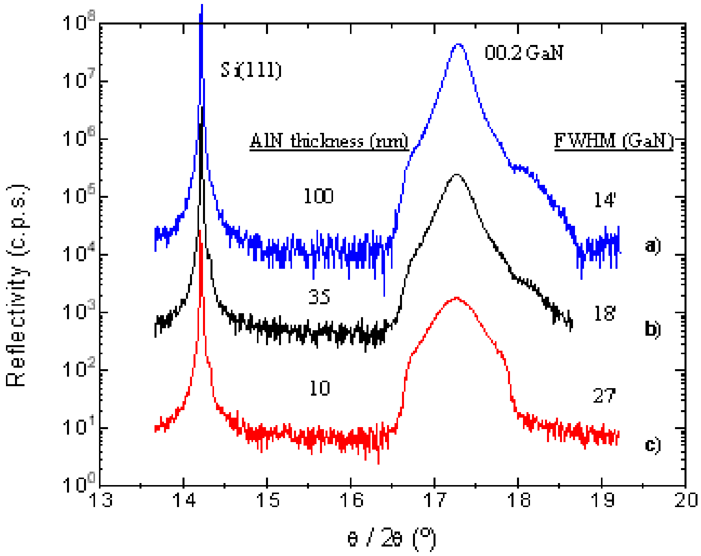

Figure 5. XRD profiles of similar GaN films grown on different thicknesses AlN buffer layers: a) 100nm of AlN, FWHM (GaN)= 14 arcmin, b) 35nm of AlN, FWHM(GaN)= 18 arcmin, and c) 10nm of AlN with FWHM (GaN) = 27 arcmin.

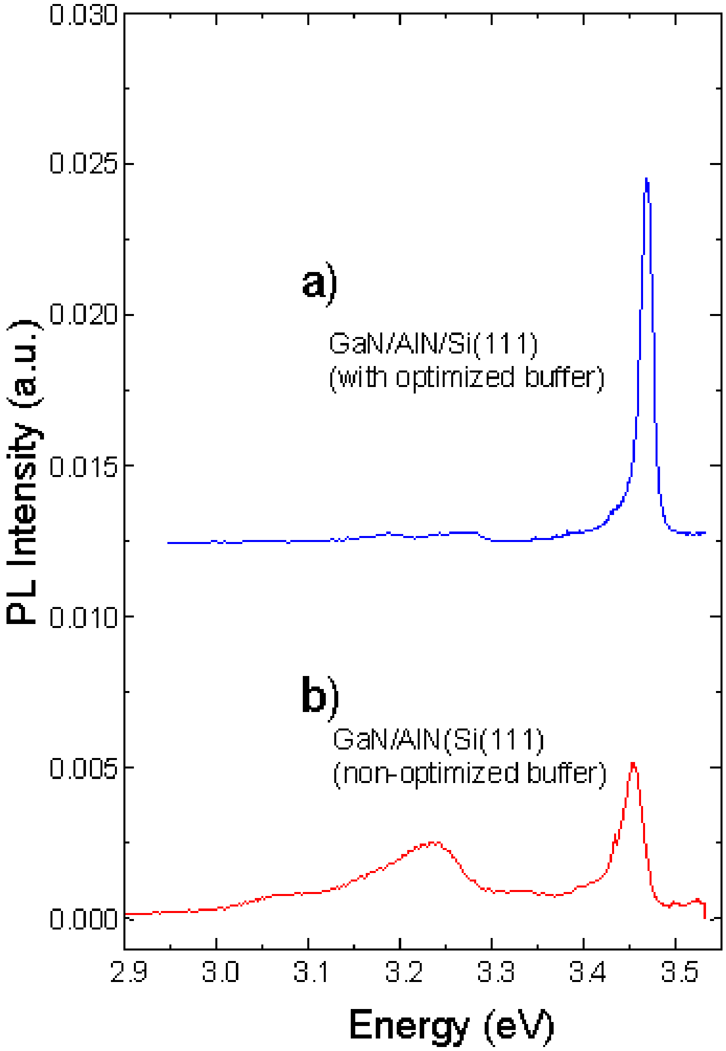

Figure 6. Low temperature photoluminescence spectra of GaN layers grown on a) an optimized and b) on a non-optimized AlN buffer layer.