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The Influence of AlN Buffer Layer Thickness on the Growth of GaN on a Si(111) Substrate with an Ultrathin Al Layer

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Abstract : It was studied the effect of a pre-deposited ultrathin Al layer as part of a buffer layer for the growth of GaN. AlN buffer layers were deposited on a Si(111) substrate using an RF sputtering technique, followed by GaN using hydride vapor phase epitaxy (HVPE). Several atomic layers of Al were deposited prior to AlN sputtering and the samples were compared with the others grown without pre-deposition of Al. And it was also studied the influence of AlN buffer layer thickness on the growth of GaN. The peak wavelength of the photoluminescence (PL) was varied with increasing the thickness of the GaN and AlN layers. The optimum thickness of AlN on a Si(111) substrate with an ultrathin Al layer was about 260Å. Scanning electron microscope (SEM) images showed coalescent surface morphology and X-ray diffraction (XRD) showed a strongly oriented GaN(0002) peak.

Key words : GaN, HVPE, AlN buffer, Si substrate

1. Introduction

GaN is an attractive material because of its potential electronic and optoelectronic device applications in the blue and ultraviolet regions. Furthermore, microstructure fabrication of these wide band gap semiconductors is important to realize high efficiency optical devices as well as quantum dots and rods[1]. However, the lack of a suitable substrate has always delayed the progress of high quality GaN epilayers. Most GaN layers are grown on sapphire substrates in spite of some serious limitations. Recently, Si(111) has been considered to be one of popular candidates as a substrate for GaN growth because of its low cost, large size and high electrical conductivity. When a Si substrate is employed for GaN growth, the use of a suitable buffer layer is essential because high quality GaN epilayers on a Si substrate have not been achieved easily due to the large lattice mismatch (17%) and the large difference in thermal expansion coefficients (56%) between GaN

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and Si. So far various types of buffer layer, such as GaAs, AlN, ZnO, 3C-SiC and AlAs have been used in the growth of GaN[2]. In particular, AlN buffer layers on a Si substrate have been shown to ease some of the difficulties of GaN growth more effectively when compared with other buffer layers[3]-[7].

The growth conditions for the AlN buffer affects strongly the growth of GaN epilayers. When GaN is grown on a Si substrate without a buffer layer, it is difficult to get high quality GaN because of the amorphous Si_xN_y layer formed by the creation of Si N bonds at the surface. In a metal organic chemical vapor deposition (MOCVD) reactor, ultrathin Al layers were deposited by exposing the Si wafer to trimethylaluminium (TMA), followed by exposure to ammonia leading to the formation of AlN. In this nitridation process Al barrier formation occurs before AlN growth. It effectively suppresses Si diffusion by forming Si N bonds[8]. Another well known phenomenon is that at high temperature, the melt back etching of Si substrates by Ga droplets tends to form Si_xN_y and leads to a rough surface. Thus, the pre deposited Al layer effectively prevents Si diffusion forming Si N bonds. In a HVPE system, the formation of an AlN layer is possible on Al_2O_3 substrates by exposing them to ammonia. However, this process is difficult in a HVPE system using Si substrates[9]. It was pre deposited the samples with several atomic layers of Al before AlN buffer layer formation and then proceeded with HVPE. This study suggests that the quality of GaN grown in a HVPE growth system is improved by employing a sputtering step to deposit ultrathin Al layers, similar to the Al layers seen in MOCVD growth[10]. And we also investigate the influence of the AlN buffer and GaN layer thickness on the optical and structural properties of GaN epilayer grown on a Al/Si(111) substrate.

2. Experimental procedure

The Si(111) substrate was cleaned chemically by using an etching solution of H_2O_2:NH_4OH:H_2O (1:1:2) and BOE (Buffered Oxide Etchant) prior to the growth[11]. At first, Al layers with few atomic thicknesses (about 5Å) were pre deposited on a Si(111) substrate by an e beam evaporator and AlN with various thicknesses was prepared by RF sputtering. The vacuum pressure of RF sputtering was 5mtorr and the plasma power was 200W. The Ar gas flow rate was 50sccm. Epitaxial growth of GaN on the Si(111) substrate was carried out in an atmospheric horizontal HVPE system. The reactor is divided into six separate temperature zones to create a definite temperature profile. The flow rate of NH_3 and HCl was kept as 300sccm and 12sccm during the growth, respectively. The growth temperature was 1050°C and Ga source temperature was maintained at 850°C[12]. GaN layers were grown from 20 minutes to 2 hours for each sample by using metallic Ga and ammonia. The temperature was maintained at 560°C for a several minutes to grow the low temperature GaN (LT-GaN) buffer layer on the AlN buffer. LT-GaN was inserted to reduce the lattice mismatch

and dislocations[13]. The samples were investigated using a PL system, SEM and XRD and compared with each other.

3. Results and discussion

It was measured the PL from the samples to characterise the optical properties. This was carried out by using a He-Cd laser (325nm) with 100-mW continuous wave as an excitation source. Fig. 1 shows the peak wavelengths of PL spectra at room temperature given as a function of the AlN thickness for GaN/AlN/Si(111). The empty circles represent the samples without an ultrathin Al layer and the solid circles, solid squares and solid triangles represent the samples with an ultrathin Al layer. Table 1 shows the experimental thicknesses of Al, AlN and GaN used.

PL peak values for GaN with a pre-deposited Al layer (solid circles) are mostly 390 nm irrespective of AlN thickness. On the other hand, for GaN without a pre deposited Al layer (empty circles) the peak value deviates from 390nm in proportion to the thickness of AlN. The inset in Fig.1 shows the full width at half maximum (FWHM) values of the PL for samples in group A.

Table 1 Thicknesses of Al, AlN, GaN used

	Al	AlN	GaN
Group A	5Å	20Å**(a)**, 50Å, 70Å	About 41μm
Group B	5Å	20Å**(b)**, 60Å	About 123μm
Group C	5Å	120Å, 180Å, 260Å**(c)**, 360Å, 500Å, 600Å	About 123μm
Group D	5Å	260Å**(d)**	About 246μm

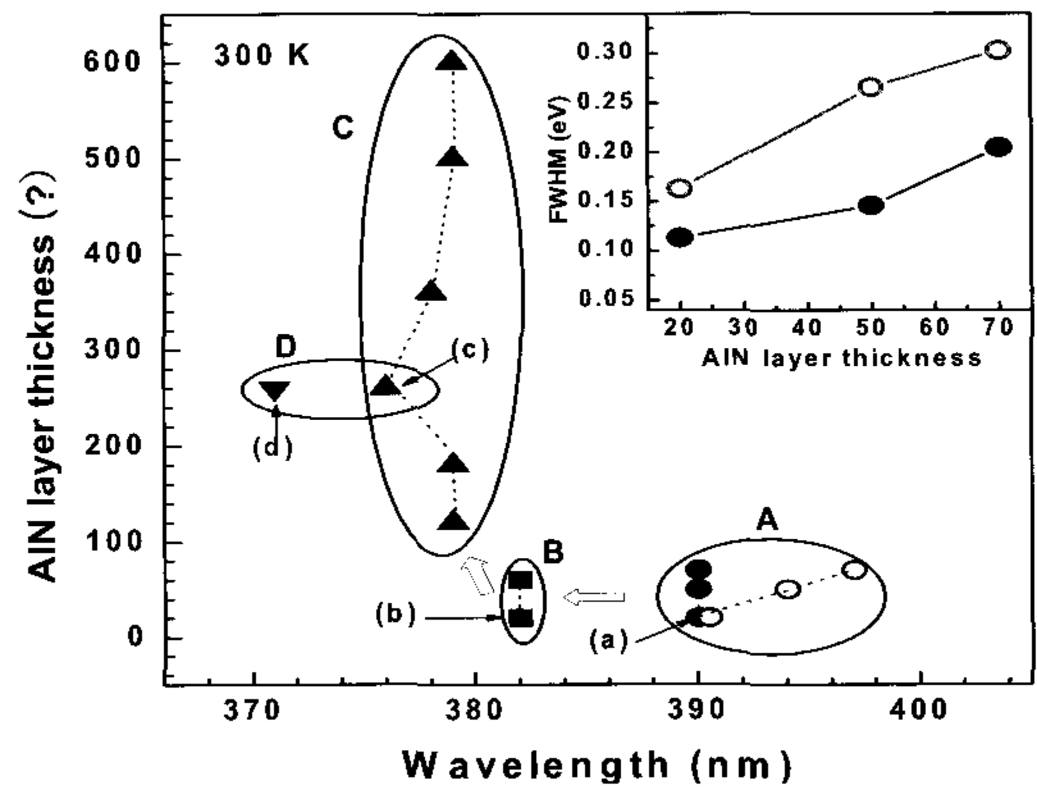


Fig. 1 Variation of PL peak wavelength with AlN and GaN layers thickness. The inset shows the FWHM values of samples in group A with a pre-deposited Al layer (solid circles) and without a pre-deposited Al layer (empty circles).

The FWHM from the GaN samples with a pre-deposited Al layer are shown to be lower than those for the GaN samples without pre deposited Al layers. This can be explained in terms of a similar process that occurs in MOCVD growth, whereby AlN nucleation is helped by pre deposition of an ultrathin Al layer which is made by exposing the substrate to TMA for few seconds. The PL peak values move from group A to group B by increasing the growth duration of HT-GaN from 20 minutes (thickness~41μm) to 1 hour (thickness~123μm). They move from group B to group C by increasing AlN thickness from 20~60Å to 120~600Å with the same HT-GaN growth duration. The HT-GaN of samples (c) and (d) was grown for 1 hour and 2 hours, respectively, with the same 260Å thick AlN layer. Other experiments were done with 100~500Å thick AlN as a buffer layer[3]-[8]. The peak of the PL spectrum moves by increasing the thickness of GaN and the optimum

AlN thickness is 260Å. Thus it is confirmed that the threading dislocations or impurity substitutions in the overgrown GaN epilayers decrease by increasing the thickness of GaN. The peak value of sample (d) in Fig. 1 has about 5nm difference from the typical value of 366 nm measured in ordinary GaN grown on Al_2O_3[6]. This means that there is still residual stress between the buffer layer and the Si substrate. XRD is required to investigate the strain and crystalline quality of the GaN epilayer. XRD in the 2 θ-scan mode is shown in Fig. 2 for samples (a), (b), (c) and (d).

The dominant diffraction peaks of samples are 34.4° and show wurtzite GaN(0002) diffraction. There are other peaks at GaN($10\bar{1}1$), GaN(102), GaN ($10\bar{1}2$), GaN($10\bar{1}3$) and GaN(0004) with 2θ = 36.6°, 47.9°, 57.6°, 63.3° and 72.7°, respec- tively. However, these peaks,

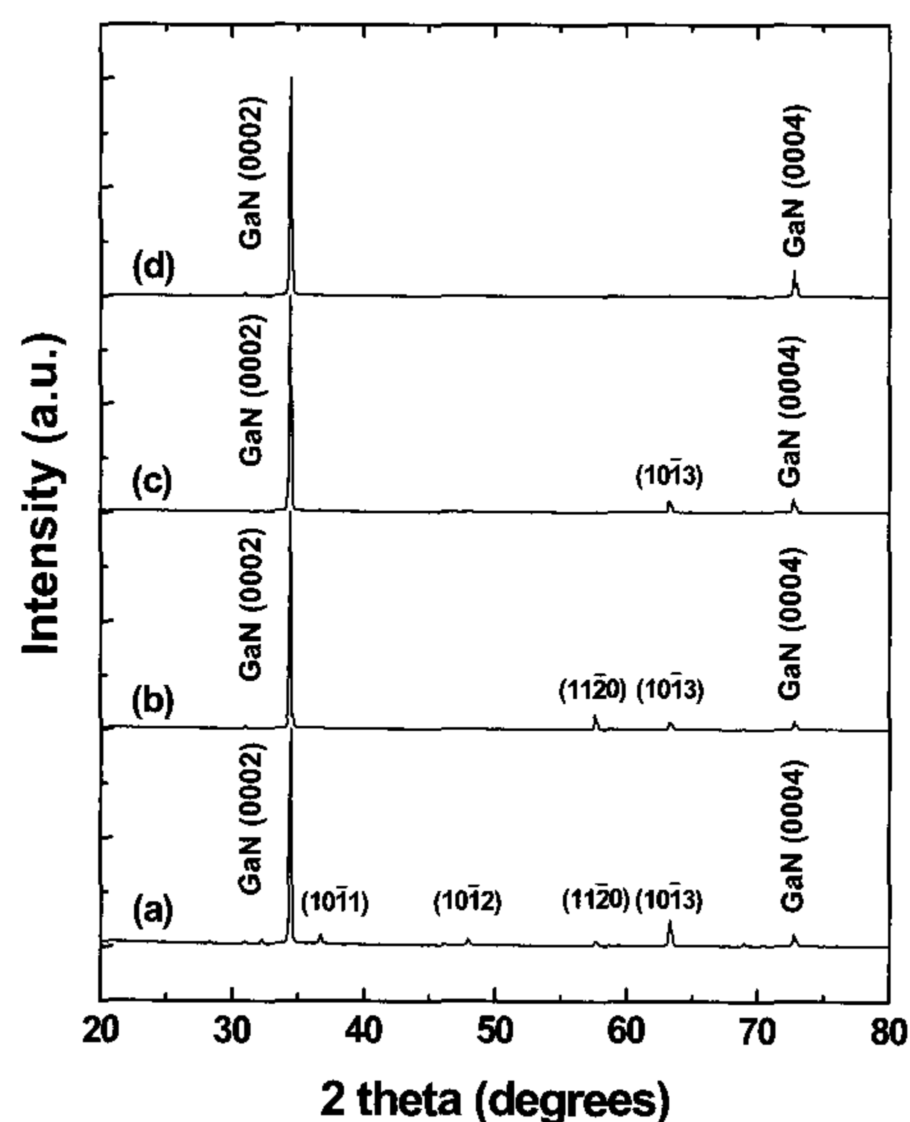
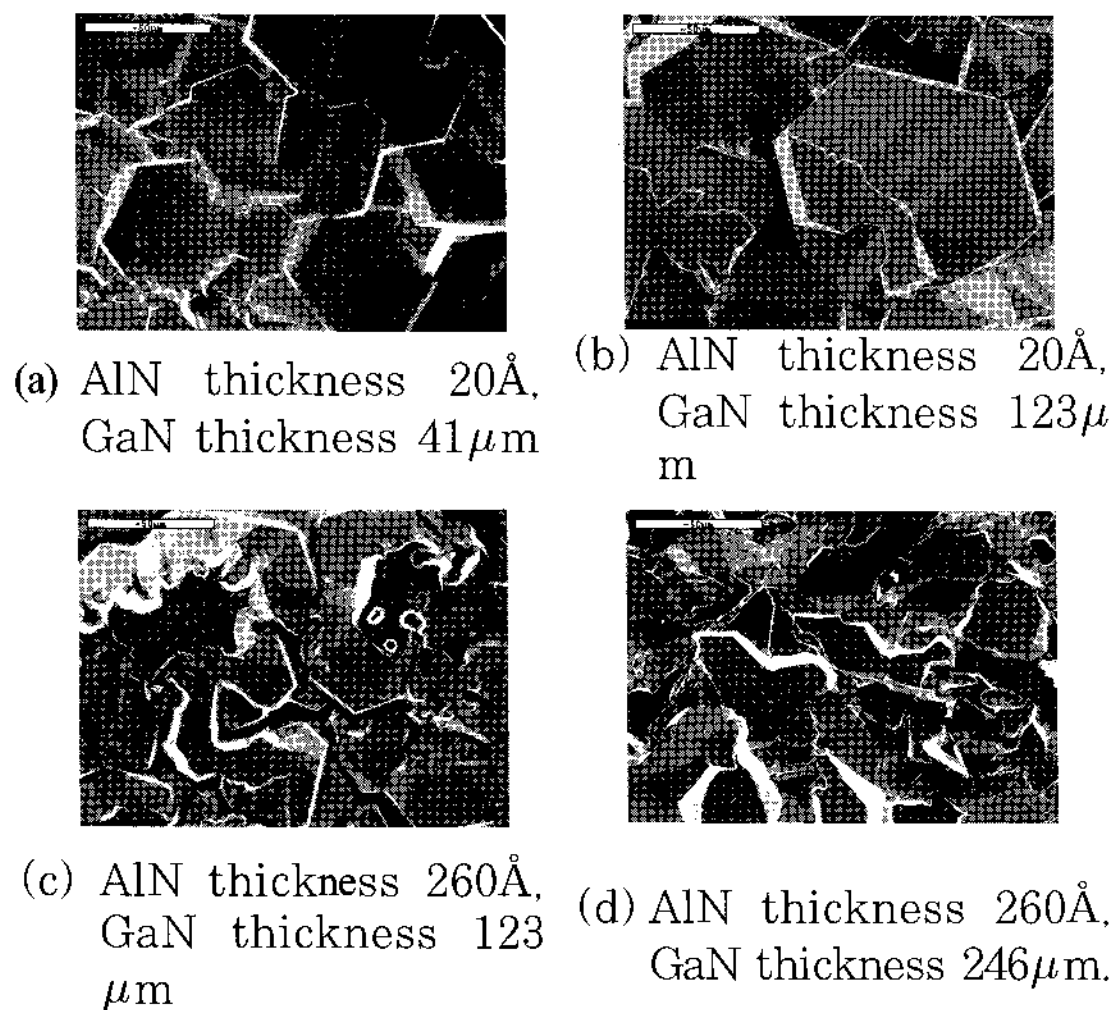


Fig. 2 X ray diffraction patterns of GaN grown on a Si(111) substrate with a LT GaN/AlN/Al buffer layer.

except (0004), disappear when moving from sample (a) to sample (d). These addi- tional weak peaks in the XRD pattern indicate the polycrystalline nature of the GaN. Such results are caused by island growth of GaN around an AlN nucleation site. It is believed that a smooth morphology is achieved by increasing the growth duration of the GaN. Thus the GaN with 2 hours growth duration and a 260 Å thick AlN buffer layer was strongly oriented along (0002) and (0004) as shown in sample (d) of Fig. 2. These results suggest that optimization of the AlN/Al buffer layer thickness could be one of the factors to adjust when determining the crystallinity of thick GaN.

It is known that a strong GaN peak appears at $2\theta=34.6°$ when there is no residual stress between the GaN layer and substrate, and the c axis length of GaN is 5.185Å[14]-[16]. However, the 2θ value for GaN (0002) is 34.4° in Fig. 2 and the GaN feels stress: the calculated thermal strain is given by $\epsilon=\Delta T(\alpha_1-\alpha_2)$ = 0.00206, where α_1 and α_2 are the thermal expansion coefficients of Si ($3.59\times10^{-6}K^{-1}$) and GaN ($5.6\times10^{-6}K^{-1}$), respectively, and ΔT is the difference between room temperature and the growth temperature. Thus the thermal stress between the GaN layer and the Si substrate is $\sigma=0.4928$ Gpa in this investigation and results in compressive stress under Ga rich conditions[17]-[19].

Fig. 3 shows SEM images of GaN/AlN/ Si(111) for different sample thicknesses (a) AlN 20Å, GaN 41μm, (b) AlN 20Å, GaN 123μm, (c) AlN 260Å, GaN 123μm and (d) AlN 260Å, GaN 246μm.



(a) AlN thickness 20Å, GaN thickness 41μm
(b) AlN thickness 20Å, GaN thickness 123μm
(c) AlN thickness 260Å, GaN thickness 123 μm
(d) AlN thickness 260Å, GaN thickness 246μm.

Fig. 3 SEM morphology of GaN on Si(111) using an AlN buffer layer pre deposited with Al

Hexagonal morphology is clearly seen in the initial growth stage of Fig. 3(a) and there are regular small pits on the surface. The hexagonally shaped GaN surfaces begin to have flat areas when the GaN growth time is increased, as shown in Fig. 3(b). The side length of the hexagonal structures is ~60μm. In Fig. 3(c), the flat areas are broadened gradually and hexagonal surface features are partially visible. It is observed in Fig. 3(d) that two dimensional growth develops at each hexagonal center and then the surface coalesces. However the surface of GaN layers contains partially a polycrystalline structure, even though XRD shows only GaN(0002) and (0004) wurtzite crystalline patterns. This means that the sample (d) preferentially has the crystalline structure oriented along the hexagonal c axis. These growth processes and the surface indices of the GaN hexagonal column islands are drawn in Fig. 4.

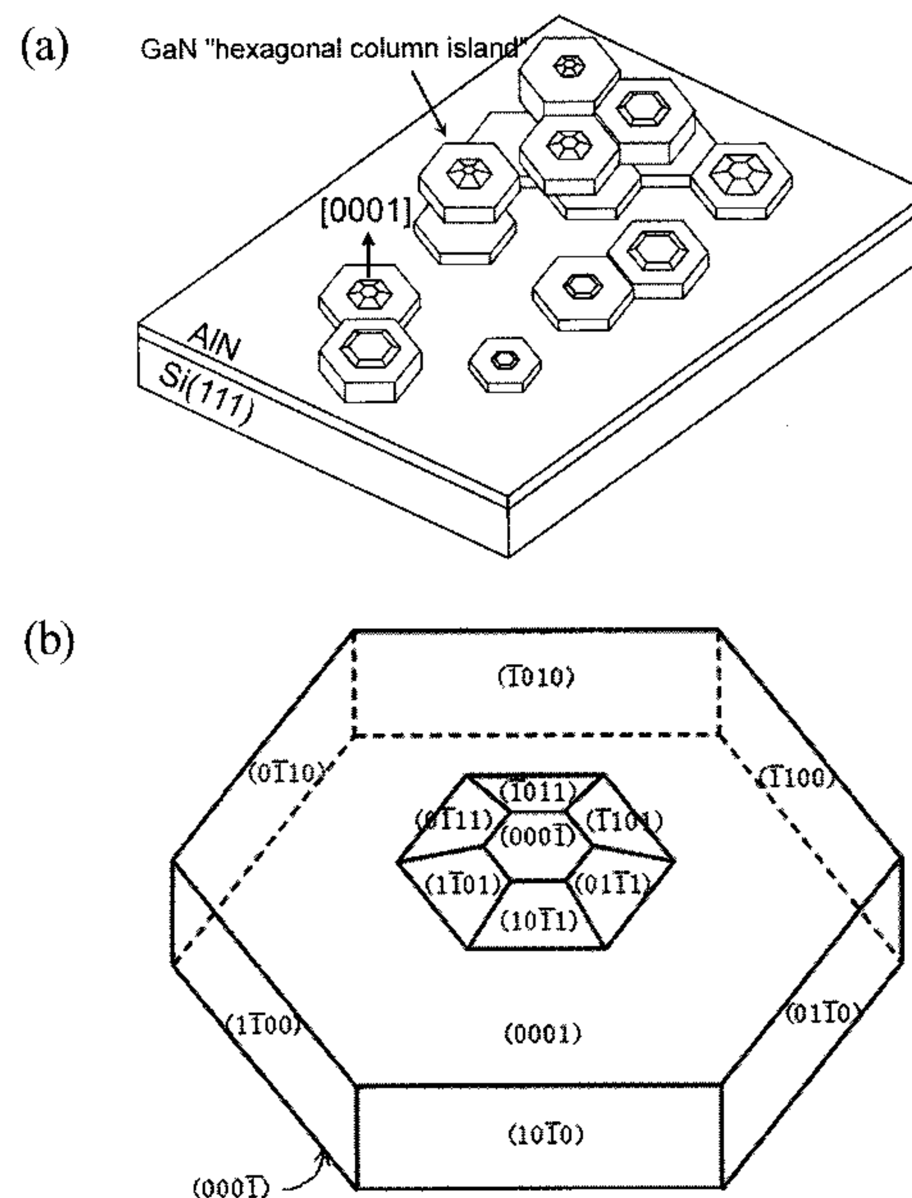


Fig. 4 Schematic diagram of GaN hexagonal column islands with 2D morphology; (a) Growth procedure picture, (b) surface indices of GaN hexagonal column islands.

The process of growth involving hexagonal units gives another viewpoint for the determination of the crystalline structure [20], [21]. In overgrowth of epitaxial GaN, the lateral to vertical growth ratio is determined by the binding energy of the growth unit. This means that the growth rate on the non polar facet {10$\bar{1}$0} is faster than that on the polar planes {0001} and {101}[22]. If {10$\bar{1}$1} planes grow faster than the {0001} planes and slower than the {10$\bar{1}$1} planes, then the microcrystal shows hexagonal column islands. However, if the {10$\bar{1}$0} planes grow faster than the {000$\bar{1}$} and {10$\bar{1}$1} planes, the morphology of the microcrystal has a hexagonal shape with inclined facets, somewhat like a small additional flat island as shown in Fig. 4(b).

4. Conclusions

In this work, we investigated that the growth behavior of GaN grown on Si(111) substrates with an ultrathin Al layer in the buffer. AlN buffer layers were prepared by RF sputtering on the ultrathin Al films. This dry deposition process can be compared with the wet deposition process of MOCVD systems using liquid Al. With that method it is easy to control the thickness of the buffer layer. It can also suppress the formation of an amorphous Si_xN_y layer effectively because the pre deposited Al layer prevents the formation of a Si N bond. We introduce a method similar in conception to the MOCVD process to grow GaN in a HVPE system. We find that a shift in PL spectra occurs by increasing the thickness of GaN and the optimum AlN thickness was ~260Å. It was observed that the surface of hexagonal structures broadened and coalesced to a two dimensional morphology by systematizing the growth conditions. The XRD pattern showed a strong peak at GaN (0002). We believe that GaN crystallinity could be improved by further optimization of the AlN/Al buffer layer. This will be studied further to improve the quality of GaN epilayers grown on Si substrates and results will be reported soon.

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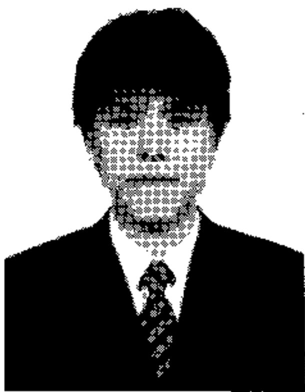

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