

High quality GaN grown on Si(111) by gas source molecular beam epitaxy with ammonia

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We describe the growth of hexagonal GaN on Si(111) by gas source molecular beam epitaxy with ammonia. The initial deposition of Al, at 1130–1190 K, resulted in a very rapid transition to a two-dimensional growth mode of AlN. The rapid transition is essential for the subsequent growth of high quality GaN and AlGaIn. This procedure also resulted in complete elimination of cracking in thick ($>2\ \mu\text{m}$) GaN layers. For layers thicker than $1.5\ \mu\text{m}$, the full width at half maximum of the (0002) GaN diffraction peak was less than 14 arc sec. We show that a short period superlattice of AlGaIn/GaN grown on the AlN buffer can be used to block defects propagating through GaN, resulting in good crystal and luminescence quality. At room temperature, the linewidth of the GaN exciton recombination peak was less than 40 meV, typical of the best samples grown on sapphire.

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There has been a number of recent attempts to prepare GaN structures on Si(111) substrates.^{1–4} However, the large difference in lattice parameters, and the strength of the Si–N bond, makes high quality epitaxy of GaN on Si quite difficult. The difficulties are alleviated by the use of a buffer layer, such as AlN, prepared first on the surface of Si,⁴ but even with such a layer most of the GaN structures grown on Si appear inferior to those prepared on more conventional substrates, such as sapphire or SiC.

Gas source molecular beam epitaxy (GSMBE) with ammonia can be used to grow high quality AlN on Si(111),⁵ and thus provide the needed buffer layer for the growth of GaN. High growth temperature of 1130–1190 K, combined with carefully controlled deposition of an Al layer at the onset of epitaxy, suppress formation of SiN_x islands and result in very rapid transition to two-dimensional (2D) growth mode of AlN.

This work describes the preparation of high quality GaN layers on Si(111) by GSMBE with ammonia. We use triple-crystal x-ray diffraction, transmission electron microscopy (TEM), Raman spectroscopy, and photoluminescence (PL) to show that these GaN structures are similar to the best samples reported on sapphire.⁶

The growth experiments were carried out on 2 and 3 in. diameter Si(111) prepared by wet chemical etching. The procedure used results in hydrogen terminated surface.⁷ Wafers prepared by this process showed the (7×7) surface structure, observed by reflection high-energy electron diffraction (RHEED), after heating to 920 K for a few minutes. The well known Si(111) surface reconstruction transition, from (7

×7) to (1×1), which occurs at $T_t=1100\text{ K}$,^{8,9} provided a convenient temperature calibration point.¹⁰

Following our previous experiments⁵ on GSMBE of AlN with ammonia, we grew AlN at $1160\pm30\text{ K}$. The process was started by exposing the Si surface to the flux of Al, estimated at $J_{\text{Al}}=(2.0\text{--}2.6)\times10^{14}\text{ at/s cm}^{-2}$, for 1–2 s, followed by 1–2 s long exposure to ammonia (with the Al flux off). This process, repeated 5–10 times, facilitated formation of oriented AlN islands on Si and suppressed formation of SiN_x islands.¹¹ The growth of AlN was started by turning on the ammonia and Al, together, with the NH₃ beam equivalent pressure of $(0.5\text{--}3)\times10^{-5}\text{ Torr}$. The Al flux was then slowly decreased, during $\sim5\text{ min}$, to $(0.8\text{--}1.3)\times10^{14}\text{ at/s cm}^{-2}$. The entire 50–120 nm AlN buffer was grown in 10–25 min. The formation of the (1×1) surface structure could be seen by RHEED after deposition of 5–7 nm of AlN. A completed 2D growth mode could be seen, by RHEED, after about 40 nm. The rapid transition to 2D growth is of interest for two reasons. First, it results in a decrease in the defect density, as shown in the TEM cross section of Fig. 1(a). Second, the rapid transition is needed to assure crack-free samples, as discussed later.

The growth of GaN was started after the AlN buffer layer completed the transition to the 2D growth. GaN layers were grown at a temperature of $1000\pm30\text{ K}$ and growth rates of $0.4\text{--}1.1\ \mu\text{m/h}$. Formation of a (2×2) surface structure could be seen after deposition of 20–100 nm of GaN, as shown in Fig. 2. The fastest 3D–2D transition in GaN was reached at the highest growth rate. This is consistent with the high value of the lateral growth rate reported for GaN at the optimal growth condition.¹² We also prepared structures incorporating short period AlGaIn/GaN superlattices (SLs) for control of defects propagating from the substrate interface.

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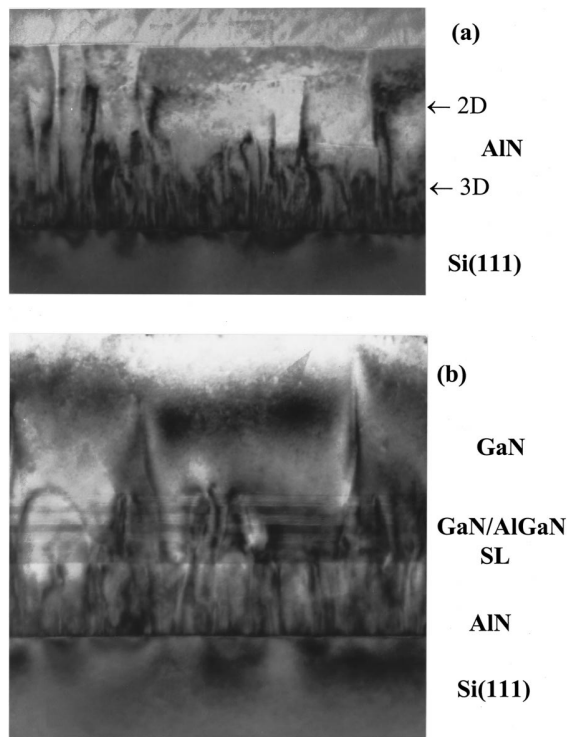


FIG. 1. TEM cross-section images: (a) of a 130-nm-thick AlN buffer layer grown on Si(111); and (b) AlGaIn/GaN SL deposited on the ~70-nm-thick AlN buffer layer. A second SL, grown on 0.4- μ m-thick layer of GaN, is not shown.

The effectiveness of such structures is demonstrated in the TEM cross section of Fig. 1(b). In addition to the SL placed at the 2D grown AlN layer, we experimented with a second SL placed on GaN growing in a 2D mode.

Figure 3 plots the x-ray diffraction linewidth, full width at half maximum (FWHM), of the (0002) peak of GaN measured as a function of the layer thickness. In samples 1.5–2 μ m thick the FWHM was as narrow as 14 arc sec. This is narrower than the values reported for GaN grown by MBE on sapphire.⁶ Figure 3 also shows the FWHM curve calculated for ideal crystalline layers as a function of thickness.¹³ A comparison of measured and calculated FWHM shows that our layers are almost completely coherent in the growth direction, for all thicknesses studied here.

X-ray diffraction measurement on a GaN structure incorporating two AlGaIn/GaN SLs is shown in Fig. 4. The structure is depicted in the inset of Fig. 4, a TEM cross section of

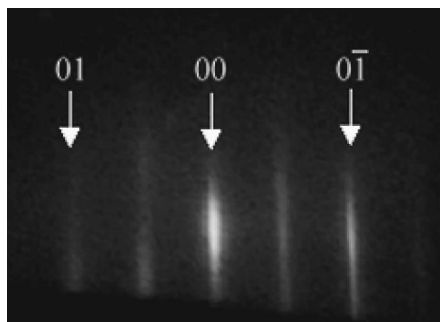


FIG. 2. The (2 $\times$ 2) RHEED pattern related to 2D growth mode of GaN. The azimuth of the electron beam is parallel to the GaN[11 $\bar{2}$ 0]||Si[110].

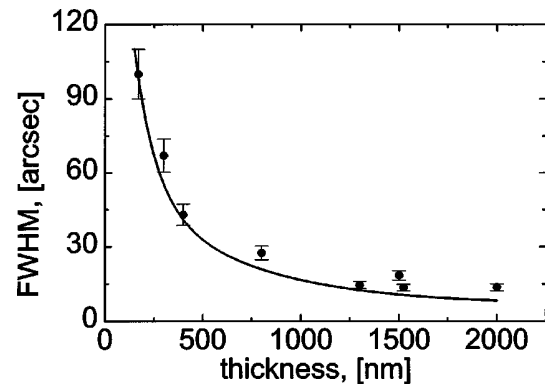


FIG. 3. X-ray linewidth (FWHM) in triple-crystal ω - 2θ scan measured for GaN layers as a function of thickness. The solid line shows the FWHM calculated for an ideal crystal (see Ref. 13).

the lower part of the structure is shown in Fig. 1(b). Figure 4 shows a narrow (0002) peak of GaN and the presence of two SLs. The sharpness of the (0002) peak and the presence of Pendellosung fringes, demonstrate high degree of top layer flatness and high crystalline quality. The coherence length calculated from the linewidth was almost equal to the thickness of the top layer of GaN.

The room temperature PL spectrum of this sample shows a free exciton (FE) peak at 3.408 eV with the FWHM of less than 40 meV, typical of the best samples grown on sapphire. Defect blocking SLs of the type shown in Fig. 4 were found important in assuring low defect (yellow) luminescence. In samples grown without SLs, the intensity of yellow luminescence was significantly stronger.

The layers of GaN grown on Si are known to crack during the cool-down cycle.¹⁴ Cracking occurs along the equivalent {1100} GaN planes and it splits the sample into small areas, with the average distance between cracks as small as 50 μ m. Cracking is believed to be an intrinsic phenomenon due to the thermal expansion coefficient mismatch between GaN and Si. However, our experience shows that cracking of GaN can be completely eliminated by suppressing formation of SiN_x islands and assuring uniform nucleation of AlN at the Si-AlN interface.¹¹

We used Raman scattering to estimate the amount of strain in the layers and to monitor the buildup of strain with the layer thickness. Raman measurements were carried out on structures similar to that illustrated in the inset of Fig. 4,

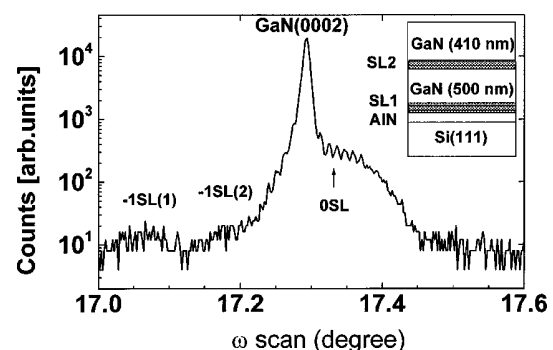


FIG. 4. X-ray diffraction data obtained on a GaN sample designed for control of crystalline quality. The presence of Pendellosung fringes (PF), near the (0002) GaN peak, is clear. The PF period corresponds to the thickness of the top GaN layer of 0.41 μ m.

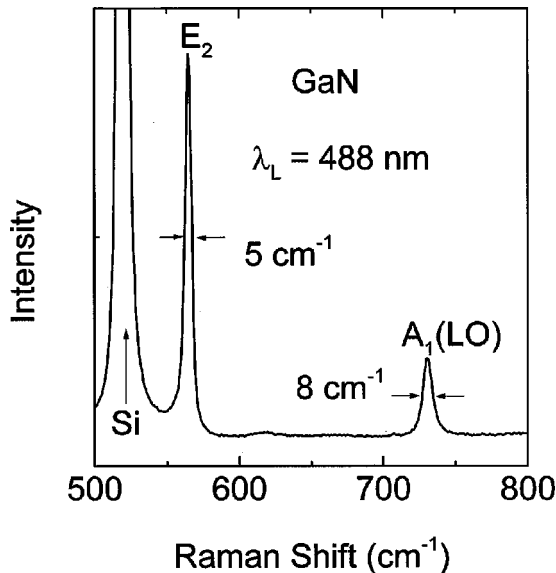


FIG. 5. Raman spectrum of a GaN sample shown in the inset of Fig. 4. The intense band at 520 cm^{-1} is arises from the Si substrate.

in which the thickness of the top GaN layer was varied between 0.4 and $2.2\text{ }\mu\text{m}$. All the measurements were done at room temperature using 488 nm , excitation from an Ar^+ laser, in a back-scattering geometry.¹⁵ Under these conditions scattering from the top GaN layer was dominant. Figure 5 shows a typical Raman spectrum, with the dominant E_2 phonon near 565 cm^{-1} . A weaker A_1 longitudinal optical band is observed at 733 cm^{-1} . This spectrum is characteristic of back scattering along the wurtzite GaN $c=(0001)$ axis. The relative intensities and FWHM are comparable to published Raman spectra of GaN on sapphire.¹⁶ We find that the E_2 band intensity is directly proportional to the film thickness. We also observe no thickness dependence to the E_2 phonon energy. Our measured value of $564.8 \pm 0.2\text{ cm}^{-1}$ implies a biaxial tensile stress of $\sim 160\text{ MPa}$ and an in-plane strain $\epsilon_a = +0.09\%$,¹⁷ independent of the sample thickness. This stress is below the ultimate strength of breaking of 400 MPa , under tensile stress, for GaN.¹⁸ Using the difference in thermal expansion coefficients for GaN and Si,¹⁹ we estimate the strain due to cooling from growth temperature to ambient at $\epsilon_a = +0.06\%$. The thermal expansion mismatch thus accounts for most of the strain present in our GaN. However,

residual stress due to lattice constant mismatch cannot be excluded.

In summary, we describe GSMBE procedures which result in high quality GaN on Si(111). The layers have excellent structural characteristics, strong and narrow room temperature PL, and are free of cracks.

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