

Growth of AlGa_N on Si(111) by gas source molecular beam epitaxy

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Gas source molecular beam epitaxy with ammonia was used to grow Al_xGa_{1-x}N on Si(111). Three types of buffer layers, containing AlN, AlGa_N/AlN, and AlGa_N/Ga_N short period superlattices, were used and their effectiveness evaluated by x-ray diffraction. We determined that a combination of AlN buffer layer, prepared under the two-dimensional growth mode, with a short period superlattice of AlGa_N/Ga_N results in the highest quality AlGa_N. Under optimized growth conditions, x-ray diffraction coherence length almost equal to the layer thickness was obtained for low Al content layers. The normalized coherence length was reduced to ~ 0.4 for $x=0.66$ and it increased again to ~ 0.75 in AlN. From room temperature band edge cathodoluminescence of AlGa_N grown on Si(111) we determined the alloy bowing coefficient of $b=1.5$ eV, in good agreement with previous results obtained by absorption measurements. © 2000 American Institute of Physics. [S0003-6951(00)03621-4]

Heterostructures of GaN and AlGa_N, grown on sapphire and SiC, are used in high mobility field effect transistors.¹⁻⁴ Structures based entirely on layers of AlGa_N, prepared on sapphire, are being developed for solar-blind photodetectors⁵⁻⁸ and are needed for light emitting diodes operating at wavelengths shorter than 360 nm. Many of these applications would benefit from AlGa_N-based heterostructures grown on Si. However, while some work has already been done to prepare AlGa_N on Si(111) by different growth methods,⁹⁻¹² the use of molecular beam epitaxy (MBE) has not been explored in much detail.¹⁰

We have recently described growth of high quality AlN and GaN on Si(111) substrates by gas source molecular beam epitaxy (GSMBE) with ammonia.^{13,14} Epitaxial layers of AlN and GaN prepared under two-dimensional growth mode exhibited excellent structural and optical properties, and were completely free of cracks. This work describes preparation of AlGa_N, over the entire range of AlN contents, with excellent crystal quality and optical properties at room temperature.

The growth experiments were carried out on 2 and 3 in. diameter wafers of exactly oriented Si(111). The wafers were prepared prior to growth as described previously.¹³ The method of substrate temperature calibration has been already described.¹³⁻¹⁵ The growth process was started by formation of an AlN buffer layer at the temperature of 1130–1190 K. This layer was nucleated by exposing the Si surface to the flux of Al, followed by a brief (1–2 s) exposure to ammonia (with the Al flux off).¹⁶ This process, repeated a few times, resulted in formation of a fully relaxed thin nucleation layer of AlN.¹⁷ Once this layer was formed, epitaxy of AlN was started by turning on the ammonia and Al and it continued until a layer thickness of 30–60 nm was reached. The (1×1) surface structure could be seen by reflection high energy electron diffraction (RHEED) and low energy electron dif-

fraction (LEED) after epitaxial deposition of 2–3 nm of AlN. The AlN buffer layer was used to inhibit formation of Si₃N₄ at the Si surface and to assure uniform nucleation of AlGa_N. In addition to this simple buffer layer between AlGa_N and the Si substrate we also experimented with two types of superlattices (SL), grown over the AlN layer. All three buffer layers are shown in a schematic drawing of Fig. 1.

In our previous experiments with GaN grown on Si(111) we used a defect blocking SL consisting of two to four pairs of Al_xGa_{1-x}N, with $x \sim 0.1-0.4$, and GaN spacers. The typical layer thicknesses were 2–4 and 3–6 nm, respectively. Two-dimensional (2D) growth mode of either GaN or AlGa_N could be reached after the first two pairs of layers, but four pairs were found to be more reliable. The efficacy of this structure in reducing the density of dislocations and crystal block boundaries propagating from the substrate–epilayer interface was confirmed by transmission electron microscopy.¹⁵ This type of a SL was also used here for the growth of Al_xGa_{1-x}N with $x < 0.4$. For AlGa_N with higher Al content we experimented with SL consisting of Al_xGa_{1-x}N and AlN pairs, with a similar range of thicknesses. The Al concentrations in the SL were equal to the Al concentrations in the main layer of AlGa_N, or slightly higher. This assures an optically transparent epitaxial struc-

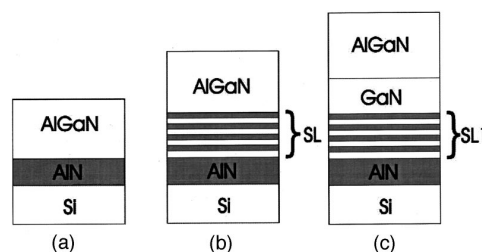


FIG. 1. Schematic cross sections indicating different buffer layers used in the preparation of AlGa_N samples grown on Si(111): (a) AlN buffer layer, (b) AlN buffer followed by a short period AlGa_N/AlN superlattice, and (c) AlN buffer followed by a short period AlGa_N/GaN superlattice and a GaN spacer layer.

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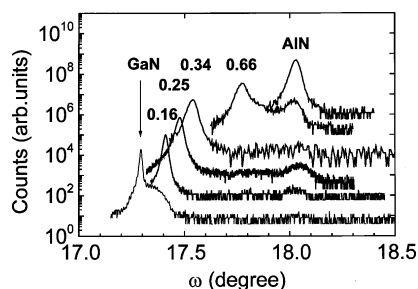


FIG. 2. High resolution x-ray diffraction of AlGa_N layers grown on Si(111) for selected compositions. The buffer structure included a thin layer of AlN followed by an AlGa_N/AlN superlattice.

ture, needed for solar-blind detectors. All the defect-blocking structures were grown at 1000 ± 30 K. AlGa_N/Ga_N SLs have been used recently in metal-organic chemical vapor deposition of GaN on Si(001).¹¹

AlGa_N layers were grown at a temperature of 1000 ± 30 K and growth rates of $0.4\text{--}1.5$ $\mu\text{m/h}$. Formation of a (2×2) surface structure could be seen reproducibly on AlGa_N grown on Al_xGa_{1-x}N/GaN superlattices. This structure is indicative of surfaces stabilized with either Al or Ga. The Al_xGa_{1-x}N with $x > 0.4$, grown on Al_xGa_{1-x}N/AlN SLs, exhibited (1×1) surface structure. The possibility of N-terminated surfaces cannot be thus excluded.

Figure 2 shows high resolution x-ray diffraction spectra of GaN, AlGa_N, and AlN layers prepared on Si(111). The ω - 2θ spectra were obtained in the double- and triple-crystal geometry. The (0002) diffraction measures crystal quality in the growth direction and is not sensitive to lateral block size, misorientation, and twist of blocks in the growth plane. The x-ray linewidth varied from ~ 20 arcsec in GaN, to ~ 130 arcsec for Al_xGa_{1-x}N with $x \sim 0.66$, and to ~ 90 arcsec in AlN.

The layers illustrated in Fig. 2 varied in thickness from 2 to 0.2 μm , making direct comparisons of x-ray linewidths unreliable. In order to correct for the sample thickness variation we calculated the coherence length, in the growth direction, for each sample. The coherence length, for each composition, was then normalized by the sample thickness. The resulting normalized coherence length is plotted in Fig. 3 as a function of the AlN concentration in the alloy. The normalized coherence length varies from almost 1 in GaN (as has been already described¹⁴), to less than 0.4 in Al_{0.66}Ga_{0.34}N, and it increases again to 0.75 in AlN. This behavior of coherence length as a function of composition could be attrib-

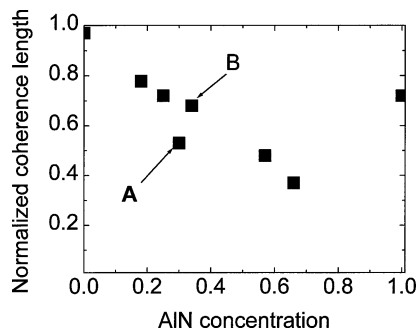


FIG. 3. Coherence length of AlGa_N in the c -axis direction, calculated from the x-ray linewidth, and normalized to the layer thickness.

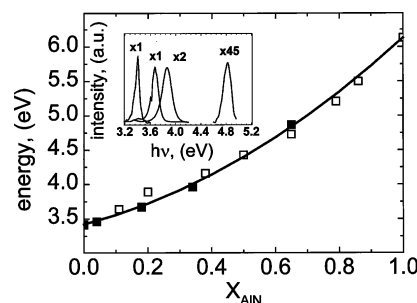


FIG. 4. Room temperature band edge CL peak position (solid square) of AlGa_N samples plotted as a function of composition. Absorption data of Brunner *et al.* Ref. 22 (open square) was used for comparison. The band gap, calculated using a bowing coefficient of 1.5 eV, is shown as a solid line. Inset shows room temperature CL spectra of selected AlGa_N samples.

uted to alloy disorder. It is known that the exciton linewidth^{18,19} and absorption edge²⁰⁻²² in AlGa_N show maximum broadening for compositions of $x = 0.6\text{--}0.8$. The exciton linewidth is proportional to $[x(1-x)]^{1/2}$,²³ a signature of the alloy disorder. We believe this effect also impacts the x-ray linewidth dependence on composition. However, the x-ray linewidth also depends on detailed growth conditions. For instance, the sample with $x = 0.30$, labeled (A), was grown directly over the AlN buffer layer. It showed a wider x-ray linewidth and therefore lower normalized coherence length of 0.53. In this sample the 2D growth mode was reached only at the end of the epitaxial process. Sample labeled (B), with a similar AlN fraction of 0.34 was grown using an AlN buffer layer, followed by a defect blocking superlattice of AlN/GaN and a GaN spacer layer. The upper part of the GaN spacer was grown in the 2D growth mode, and formation of the (2×2) surface structure was observed by RHEED. This allowed for 2D growth of the AlGa_N layer, resulting in the normalized coherence length of 0.68.

Optical properties of AlGa_N samples are illustrated in Fig. 4. The inset shows room temperature cathodoluminescence (CL) spectra of selected samples with different alloy composition. The samples were excited with a 15 keV electron beam, at a current of 0.05 mA. The excitation power density was estimated at ~ 20 W/cm². Under these conditions our samples showed band-edge and weak defect emission, similar to the “yellow line” emission of GaN. The peak wavelength of the band-edge emission varied from 3.406 eV (364.0 nm) for GaN to 4.826 eV (256.9 nm) for Al_{0.66}Ga_{0.34}N. We also show a plot of the band-edge energy as a function of the AlN content. In addition to the CL data obtained on our samples, Fig. 4 also shows the absorption data obtained on AlGa_N samples grown by MBE on sapphire.²² The two sets of samples show the same band gap dependence on composition, described by $E_g(x) = 3.42 + 1.21x + 1.5x^2$. This result is consistent with the low carrier concentration in our layers. Hall and voltage-capacitance (C - V) show electron concentrations in the range $10^{16}\text{--}10^{17}$ cm⁻³. The bowing coefficient $b = 1.5$ eV is close to that obtained previously for AlGa_N prepared on different substrates, by a variety of growth methods.^{18,21,22,24,25} This value of b is larger than that predicted from the lattice constant difference between GaN and AlN and may be due to a large scattering potential for electrons in this alloy system.²⁵

In summary, we describe the growth of high quality Al-GaN on Si(111) by GSMBE with ammonia. Best results were obtained using buffer structures consisting of a layer of AlN and an AlGaIn/GaN SL. X-ray diffraction and room temperature CL measurements show evidence of composition dependent alloy disorder.

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- ¹L. Eastman, J. Burm, W. Schaff, M. Murphy, K. Chu, H. Amano, and I. Akasaki, MRS Internet J. Nitride Semicond. Res. **1**, paper 4 (1996).
- ²D. Kuksenkov, H. Temkin, R. Gaska, and J. W. Yang, IEEE Electron Device Lett. **19**, 222 (1998).
- ³D. Kuksenkov, G. E. Guidice, H. Temkin, and I. Adesida, Electron. Lett. **34**, 2274 (1998).
- ⁴T. Egawa, H. Ishikawa, M. Umeno, and T. Jimbo, Appl. Phys. Lett. **76**, 121 (2000) and references therein.
- ⁵B. W. Lim, Q. C. Chen, J. Y. Yang, and M. A. Khan, Appl. Phys. Lett. **68**, 3761 (1996).
- ⁶A. Osinsky, S. Gangopadhyay, B. W. Lim, M. Z. Anwar, M. A. Khan, D. Kuksenkov, and H. Temkin, Appl. Phys. Lett. **72**, 742 (1998).
- ⁷G. Parish, S. Keller, P. Kozodoy, J. P. Ibbetson, H. Marchand, P. T. Fini, S. B. Fleischer, S. P. DenBaars, U. K. Mishra, and E. J. Tarsa, Appl. Phys. Lett. **75**, 247 (1999).
- ⁸F. Omnès, N. Marengo, B. Beaumont, Ph. de Mierry, E. Monroy, F. Calle, and E. Muñoz, J. Appl. Phys. **86**, 5286 (1999) and references therein.
- ⁹S. Guha and N. A. Bojarzuk, Appl. Phys. Lett. **72**, 415 (1998).
- ¹⁰M. A. Sánchez-García, E. Calleja, F. B. Narajo, F. J. Sánchez, F. Calle, E. Muñoz, A. M. Sánchez, F. J. Pacheco, and S. I. Molina, J. Cryst. Growth **201/202**, 415 (1999).
- ¹¹X. Zhang, S.-J. Chua, P. Li, K.-B. Chong, and Z.-C. Feng, Appl. Phys. Lett. **83**, 1984 (1999).
- ¹²H. Ishikawa, G.-Y. Zhao, N. Nakada, T. Egawa, T. Jimbo, and M. Umeno, Jpn. J. Appl. Phys., Part 2 **38**, L492 (1999).
- ¹³S. A. Nikishin, V. G. Antipov, S. Francoeur, N. N. Faleev, G. A. Seryogin, V. A. Elyukhin, H. Temkin, T. I. Prokofyeva, M. Holtz, A. Konkar, and S. Zollner, Appl. Phys. Lett. **75**, 484 (1999).
- ¹⁴S. A. Nikishin, N. N. Faleev, V. G. Antipov, S. Francoeur, L. Grave de Peralta, G. A. Seryogin, H. Temkin, T. I. Prokofyeva, M. Holtz, and S. N. G. Chu, Appl. Phys. Lett. **75**, 2073 (1999).
- ¹⁵S. A. Nikishin, N. N. Faleev, H. Temkin, and S. N. G. Chu, *State-of-the-Art Program on Compound Semiconductors (SOTAPOCS XXXI)*, Honolulu, Hawaii, October 17–22, 1999 (Electrochemical Society, New York, 1999), Vol. 99-17, p. 238.
- ¹⁶S. A. Nikishin and H. Temkin (unpublished).
- ¹⁷S. A. Nikishin, N. N. Faleev, V. G. Antipov, S. Francoeur, L. Grave de Peralta, G. A. Seryogin, M. Holtz, T. I. Prokofyeva, S. N. G. Chu, A. S. Zubrilov, V. A. Elyukhin, I. P. Nikitina, A. Nikolaev, Yu. Melnik, V. Dmitriev, and H. Temkin, paper number W8.3 in Proceedings of 1999 Fall MRS Meeting (unpublished).
- ¹⁸M. R. H. Khan, Y. Koide, H. Itoh, N. Sawaki, and I. Akasaki, Solid State Commun. **60**, 509 (1986).
- ¹⁹A. S. Zubrilov, D. V. Tsvetkov, V. I. Nikolaev, and I. P. Nikitina, Semiconductors **30**, 1069 (1996).
- ²⁰G. A. Korkotashvili, A. N. Pikhtin, I. G. Pichugin, and A. M. Tsaregorodtsev, Sov. Phys. Semicond. **18**, 913 (1984).
- ²¹M. Stutzmann, O. Ambacher, A. Cros, M. S. Brandt, H. Angerer, R. Dimitrov, N. Reinacher, T. Metzger, R. Hopler, D. Brunner, F. Freudenberger, R. Handschuh, and Ch. Deger, Mater. Sci. Eng., B **50**, 212 (1997).
- ²²D. Brunner, H. Angerer, E. Bustarret, F. Freudenberger, R. Höpler, R. Dimitrov, O. Ambacher, and M. Stutzmann, J. Appl. Phys. **82**, 5090 (1997).
- ²³E. F. Schubert, E. O. Gobel, Y. Horikoshi, K. Ploog, and H. J. Queisser, Phys. Rev. B **30**, 813 (1984).
- ²⁴J. Hagen, D. R. Metcalfe, D. Wickenden, and W. Clark, J. Phys. C **11**, L143 (1978).
- ²⁵A. S. Zubrilov, Yu. V. Melnik, A. E. Nikolaev, D. V. Tsvetkov, V. V. Tretyakov, M. A. Jakobson, D. K. Nelson, and V. A. Dmitriev, Abstracts of 2nd Russian Workshop Gallium Nitride, Indium Nitride, Aluminum Nitride: Structures and Devices, June 2, 1998, St. Petersburg, Russia, p. 34 [in Russian].