Effect of H2 Carrier Gas Flow Rate during AlN Growth on Epitaxial GaN

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Abstract

Abstract: AlN and GaN thin films were grown on Si(111) substrates by metalorganic chemical vapor deposition (MOCVD). The influence of carrier gas (H2) flow rate variations during AlN buffer layer growth on GaN epitaxial layers was investigated using high-resolution X-ray diffraction, spectroscopic ellipsometry, and atomic force microscopy. Spectroscopic ellipsometry measurements revealed that the thickness of AlN increased with increasing H2 flow rate for the same growth duration, indicating that higher H2 flow rates enhance the AlN growth rate. Atomic force microscopy analysis demonstrated that the surface roughness of AlN also increased with increasing H2 flow rate. Characterization of GaN showed that as the H2 flow rate during AlN growth increased, the full width at half maximum (FWHM) of GaN' s (0002) and (10-12) peaks increased, corresponding to increased screw-type and edge-type threading dislocation densities. This may be attributed to the larger thickness of the AlN buffer layer, leading to degraded crystal quality of GaN. The experimental results indicate that using a lower H2 flow rate for AlN buffer layer growth can control the AlN growth rate and, to a certain extent, help improve the crystal quality of GaN.

Full Text

Influence of H2 Carrier Gas Flow During AlN Buffer Growth on Epitaxial GaN

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Abstract

AlN buffer layers and GaN epitaxial films were grown on Si(111) substrates by metalorganic chemical vapor deposition (MOCVD). The influence of H2 carrier gas flow rate during AlN buffer layer growth on the GaN epitaxial layer was investigated using high-resolution X-ray diffraction (HRXRD), spectroscopic ellipsometry, and atomic force microscopy (AFM). Ellipsometry measurements revealed that AlN thickness increased with H2 flow rate under identical growth durations, indicating that higher H2 flow enhances AlN growth rate. AFM characterization showed that AlN surface roughness also increased with H2 flow rate. Analysis of the GaN layers demonstrated that increasing H2 flow during AlN growth led to broader full width at half maximum (FWHM) values for both GaN (0002) and (10-12) diffraction peaks, corresponding to increased densities of screw and edge threading dislocations. This degradation in GaN crystal quality likely stems from the greater thickness of the AlN buffer layer. The experimental results demonstrate that using a lower H2 flow rate for AlN buffer growth can control the growth rate and thereby improve GaN crystal quality.

Keywords: GaN; AlN buffer; carrier gas; Si substrate; MOCVD
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Introduction

Third-generation semiconductors such as GaN offer numerous advantages including wide bandgap, high breakdown field, high thermal conductivity, and corrosion resistance. In particular, AlGaN/GaN heterostructures exhibit high-density, high-mobility two-dimensional electron gas (2DEG), enabling widespread applications of GaN-based microelectronic devices in both military and civilian sectors. For military applications, GaN devices can operate at temperatures ranging from 600–1100°C and offer superior high-frequency, high-power, and radiation-hardened performance, making them highly attractive for aerospace and defense systems. As GaN device technology matures, increased adoption in aerospace systems will maximize operational capability and reliability. In civilian applications, the high-frequency and high-power handling capabilities of GaN-based devices are crucial for developing amplifiers, modulators, and other key components in advanced communication networks.

One factor limiting GaN device quality and cost is the substrate material. While SiC substrates are commercially available, they remain expensive. Sapphire substrates suffer from poor electrical conductivity, limiting their applicability. In contrast, Si substrates are inexpensive and enable potential integration with existing Si-based electronics, making GaN growth on Si(111) a widely studied approach with steadily improving crystal quality. However, Si substrates exhibit large lattice mismatch (17%) and thermal mismatch (56%) with GaN, causing cracks to appear in GaN epitaxial layers beyond a critical thickness. To address this, researchers have investigated various buffer layers for high-quality GaN epitaxy on Si. Low-temperature GaN (LT-GaN) buffers, commonly used on sapphire and SiC, are unsuitable for Si because the high-temperature GaN growth process promotes LT-GaN decomposition, and Ga reacts with Si to form alloys that roughen the substrate surface and degrade crystal quality. Alternative buffers such as porous Si, SiNx, 3C-SiC, AlAs, GaAs, -Al2O3, and AlN have been explored. Among these, AlN is particularly effective because Al-N bonds form more readily than Si-N bonds, suppressing polycrystalline SiNx formation and improving GaN crystal quality. Additionally, AlN buffer layers offer better thermal stability than LT-GaN buffers, making them the standard choice for GaN epitaxy on Si.

In the two-step epitaxial growth process, the AlN buffer layer significantly influences the subsequent GaN epitaxial layer. B.S. Zhang et al. found that increasing AlN buffer thickness enhanced surface roughness due to a stress transition from compressive to tensile, which suppresses Al adatom migration and promotes vertical over lateral growth. Zhao Degang et al. demonstrated that for 20 nm AlN buffers, appropriate annealing time promoted lateral growth and island coalescence during initial GaN growth. When AlN surface island density was high and grain size small, GaN coalesced quickly but contained numerous dislocations; conversely, lower island density and larger grain size yielded better GaN crystal quality. Previous studies also examined relationships between AlN growth rate and temperature, chamber pressure, and NH3 flow, finding that lower temperatures and reduced pressure enhanced growth rate and Al incorporation in AlGaN. This work specifically investigates the effect of H2 carrier gas flow rate during AlN growth on the resulting GaN epitaxial layer quality using MOCVD, HRXRD, AFM, and spectroscopic ellipsometry.

Experimental Details

GaN layers were grown using a Veeco D180 MOCVD system on 2-inch (5.08 cm) n-type Si(111) substrates with resistivity of 1–15 Ω · cm and thickness of 430 m. Prior to growth, substrates underwent a multi-step cleaning process to remove surface contaminants: 8-minute ultrasonic cleaning in acetone, followed by 8 minutes in ethanol to eliminate organic contaminants; cleaning in Solution 1 (NH3 · H2O:H2O2:H2O) to remove acidic contaminants; and cleaning in Solution 2 (HCl:H2O2:H2O = 1:2:7) to remove alkaline contaminants. Immediately before loading into the reactor, substrates were rinsed in dilute HF solution. Trimethylgallium (TMGa) and trimethylaluminum (TMAl, formula: Al(CH3)3) served as Ga and Al sources, respectively, while high-purity ammonia (NH3) provided the N source. No intentional doping was performed. Substrate cleaning concluded with a 10-minute thermal treatment at 1100°C.

Two sample series were prepared. The first series examined AlN buffer layers grown with different H2 flow rates, yielding thicknesses of 108 nm, 208 nm, 266 nm, and 269 nm at H2 flows of 8,000 sccm, 15,000 sccm, 18,000 sccm, and 30,000 sccm, respectively. All other growth parameters remained constant: temperature 950°C, pressure 30 torr, TMAl flow 76 mol/min, NH3 flow 3,000 sccm, and growth time 40 minutes. AlN buffer thickness was measured using an MD2000D spectroscopic ellipsometer, and surface morphology was characterized with a Veeco Dimension 3100 AFM.

The second series comprised GaN samples grown on AlN buffers prepared at different H2 flow rates. Following AlN buffer deposition, GaN epitaxial layers approximately 1.2 m thick were grown using identical processes. The H2 flows during AlN growth were 16,000 sccm, 20,000 sccm, and 24,000 sccm for samples A, B, and C, respectively. All other AlN growth parameters matched the first series. After growth, GaN crystal quality was evaluated by double-crystal -scan using a Bruker D8 Discover HRXRD system. The FWHM values of (0002) and (10-12) diffraction peaks obtained from -scans were used to characterize GaN crystal quality.

Results and Discussion

[Figure 1: see original paper] shows the relationship between AlN buffer layer growth rate and H2 flow rate. For H2 flows of 8,000 sccm, 15,000 sccm, 18,000 sccm, and 30,000 sccm, the corresponding growth rates were 2.7 nm/min, 5.2 nm/min, 6.6 nm/min, and 6.7 nm/min, respectively, demonstrating that AlN growth rate increases with H2 flow rate. Besides the primary reaction between Al(CH3)3 and NH3 to form AlN and CH4, parasitic reactions can occur as shown in equations (1)-(5) [13]. During MOCVD growth, dimer [Al(CH3)2NH2]2 and trimer [Al(CH3)2NH2]3 species formed via reactions (4) and (5) produce AlN particles that are swept away by the gas flow without contributing to growth [13]. In this study, reduced H2 flow increases the probability of encounters between NH3 and Al(CH3)3 molecules. Additionally, lower carrier gas flow reduces its partial pressure, causing the partial pressures of NH3 and Al(CH3)3 to increase correspondingly to maintain constant total pressure, thereby promoting prereaction between the two precursors [15].

[Figure 2: see original paper] illustrates the dependence of AlN buffer layer surface roughness on H2 flow rate. At H2 flows of 8,000 sccm, 15,000 sccm, and 18,000 sccm, the surface roughness values were 6.73 nm, 6.49 nm, and 9.39 nm, respectively, indicating that higher H2 flow tends to increase AlN surface roughness. This trend likely relates to increased AlN buffer thickness. Previous studies show that AlN buffer layers initially experience compressive stress during epitaxy on Si, which gradually transitions to tensile stress [4]. Compressive stress facilitates adatom migration, whereas tensile stress reduces Al adatom mobility [4]. Under tensile stress, lateral growth of AlN is suppressed while vertical growth rate relative to the surface increases, resulting in greater roughness. [Figure 3: see original paper] presents three-dimensional surface morphologies of AlN buffers grown at different H2 flows over a 2 m × 2 m scan area. As AlN buffer thickness increases, surface island density rises and islands become sharper, indicating faster vertical growth. Although island size increases with thickness, this differs from previous observations [4] because those studies varied growth time to control thickness, whereas this work uses H2 flow to modulate growth rate. Comparing the cases of 8,000 sccm and 15,000 sccm H2, the 18,000 sccm condition yields faster AlN growth. [Figure 4: see original paper] shows two-dimensional surface morphologies of AlN buffers grown at different H2 flows. Rapid island coalescence produces larger grain sizes but poorer island orientation, as evident in [FIGURE:4(c)].

[Figure 5: see original paper] shows the FWHM values of (0002) and (10-12) diffraction peaks for samples A, B, and C. For the (0002) reflection, the FWHM values are 681 arcsec, 746 arcsec, and 763 arcsec for samples A, B, and C, respectively. For the (10-12) reflection, the corresponding values are 728 arcsec, 709 arcsec, and 787 arcsec. The FWHM of (0002) XRD reflects the density of screw and mixed threading dislocations, while the FWHM of (10-12) reflects the density of edge and mixed threading dislocations [17]. These results indicate that increasing H2 flow during AlN growth elevates both screw and edge threading dislocation densities in the GaN epitaxial layer. The first sample series confirmed that higher H2 flow increases AlN growth rate and thickness while increasing island density. Therefore, as H2 flow increases from 16,000 sccm to 24,000 sccm, samples A, B, and C exhibit sequentially increasing AlN thickness and likely higher island density. Since GaN nucleates on top of AlN islands [16], higher AlN island density provides more GaN nucleation sites. GaN islands grow in a three-dimensional mode [16], and high-density AlN islands promote rapid coalescence during initial GaN growth, generating numerous edge threading dislocations [12]. Furthermore, high H2 flow during AlN growth produces poorly oriented islands, which degrades GaN island orientation and increases screw threading dislocation density. The experimental results demonstrate that an AlN buffer grown at 30 torr, NH3 = 3,000 sccm, and H2 flow of 16,000 sccm optimizes GaN crystal quality.

This study investigated the influence of H2 carrier gas flow rate during AlN buffer growth on GaN epitaxial layers. Increasing H2 flow enhances AlN growth rate and surface roughness. The tensile stress in AlN buffers suppresses Al adatom surface migration, strengthening vertical growth relative to the surface. Consequently, AlN buffers grown at higher rates exhibit greater surface roughness within the same growth time. Moreover, increasing H2 flow during AlN growth degrades GaN crystal quality due to high-density island formation on the AlN surface, which promotes rapid GaN coalescence during initial growth, and poor island orientation, which generates numerous dislocations. The results demonstrate that using a lower H2 flow rate for AlN buffer growth can improve GaN crystal quality.

References

[1] Li Y Z, Xing Y H, Han J, etal. Investigation of Non-doped GaN Grown on Sapphire Substrate [J]. Chin. J. Lumin.2012, 33(10): 1084-1087(in Chinese)

[2] Xing Y H, Han J, Deng J, etal. Properties of the InGaN: Mg with roughened surface and high hole concentration and its light-emitting diode application [J]. Journal of Optoelectronics · Laser，2011,22(5):666-668(in Chinese)

[3] Srinivasan R, Redwing J M. Growth stresses and cracking in GaN films on (111)Si grown by metal-organic chemical-vapor deposition. I. AlN buffer layers [J]. Appl. Phys. Lett,2005, 98:0235141-0235149

[4] Zhang B S, Wu M, Shen X M, etal. Influence of high-temperature AIN buffer thickness on the properties of GaN grown on Si(111)[J]. J. Cryst. Growth, 2003, 258(1):34-40

[5] Matoussi A, Boufaden T, Missaoui A. Porous silicon as an intermediate buffer layer for GaN growth on (100) Si[J]. Microelectron. J, 2001, 32(12) :995-998.

[6] Tamura M, López-lópez M, Yodo T. Effects of the substrate tilting angle on the molecular beam epitaxial growth of GaAs on Si(110)[J]. J. Vac. Sci. Technol. B, 2000, 19(4):1567-1571

[7] Sanchez-Garcia M A, Ristic J, Calleja E, etal. Characterization of GaN quantum discs embedded in AlxGa1-xN nanocolumns grown by molecular beam epitaxy [J] Phys. State Sol. A, 2002, 68(12):192-198

[8] Strittmatter A, Bimberg D, Krost A, etal. Structural investigation of GaN layers grown on Si(111) substrates using a nitridated AlAs buffer layer[J] J. Crystal Growth, 2000, 221(1):293-296.

[9] Yang J W, Sun C J, Chen Q, etal. High quality GaN-InGaN heterostructures grown on (111) silicon substrates [J] Appl. Phys. Lett, 1996, 69(23):3566-3568.

[10] Kobayashi N P, Kobayashi J T, Dapkus P D, etal. GaN growth on Si(111) substrate using oxidized AlAs as an intermediate layer[J] Appl. Phys. Lett,1997,71(24):3569-3571.

[11] Watanabe A, Takeuchi T, Hirosawa K. Growth of single crystalline GaN on a Si substrate using AlN as an intermediate layer [J] J. Cryst. Growth, 1993, 128(1):391-

[12] Zhao D G, Zhu J J, Liu Z S, etal. Surface morphology of AlN buffer layer and its effect on GaN growth by metal organic chemical vapor deposition [J]. Appl. Phys. Lett. 2004, 85(9):1499-1501

[13] Zhao D G, Zhu J J, Jiang D S, etal. Parasitic reaction and its effect on the growth deposition[J]. J.Cryst.Growth.2006,289(1):72-75 bymetalorganic chemical vapor

[14] Theodoros G, Mihopoulos, Gupta V,etal. A reaction transpory model for AlGaN MOVPE growth [J] J.Cryst.Growth, 1998, 195(1): 733-739.

[15] Lu D Q, Zhang R, Xiu X Q, etal. Influence of Carrier Gas Flow Rate on the Optical Propertiesof GaN Films Grown by HVPE [J]. Research & Progress of SSE. 2002,22(4):385-390(in Chinese)

[16] Krost, Dadgar, Blasing. Evolution of stress in GaN heteroepitaxy on AlN/Si(111): From hydrostatic compressive tensile[J] Appl. Phys. Lett, 2004, 85(16):3441-3443 to biaxial

[17] Chen Y，Wang W X，Li Y，etal. High quality GaN layers grown on SiC substrates with AlN buffers by metal organic chemical vapor deposition [J] Chin.J.Lumin. 2011，32(9): 896-901 (in Chinese)