Structural and Spectroscopic Properties of AlN Layers Grown by MOVPE

Sarad Bahadur Thapa

The effects on surface morphology and crystal quality of undoped bulk AlN layers, grown on c-plane sapphire substrates, due to the changes in growth conditions in LP-MOVPE are discussed here. The optimized growth process has led to an almost flat surface morphology with a significantly reduced number of hexagonal pits and good crystalline quality having a rms value of roughness of 0.4 nm and a FWHM value of the X-ray rocking curve for the (0002) reflection of 200 arcseconds. These excellent data were further confirmed by a strong donor-bound exciton luminescence signal with a FWHM of approx. 25 meV. Besides the structural and spectroscopic properties of AlN bulk layers, the HRXRD and TEM studies of AlN/GaN superlattice structure are reported here.

1. Introduction

Due to its wide direct band gap (approx. 6 eV) and the associated outstanding chemical and thermal stability, the realization of high-quality AlN epitaxial layers can widely extend the application field of III-V nitride materials. AlN could be a very promising material for high-temperature high-power applications, e.g. high-power field effect transistors, and opto-electronic devices, particularly UV diodes. Furthermore, AlN/GaN superlattice structures can be used in optical devices operating at telecommunication wavelengths by exploiting intersubband transitions between bound quantum well states [1].

However, the growth of AlN having excellent crystalline quality, smooth surface morphology, and good electrical and optical properties - as required for device fabrication - is always a big challenge. One major problem during the epitaxial growth of AlN is the parasitic reaction between ammonia and the metalorganic aluminum source TMAI (trimethylaluminum), which can be minimized by reducing the reactor pressure and upstream positioning of the substrate on the susceptor [2]. To obtain good crystalline quality and smooth surface morphology of AlN layers, many experiments have been carried out in low-pressure metal organic vapor phase epitaxy (LP-MOVPE) by changing the growth parameters. The effects of the variation of some basic growth parameters on the surface morphology, crystal quality, growth rate and consequently on the cathodoluminescence spectra are reported here.

2. Experiment

Undoped layers of AlN, approximately 500 nm thick, were grown on three different orientations of c-plane sapphire substrates, namely exactly oriented (on-axis), 0.3° miscut
towards m-plane (off-axis to m-plane), and 0.3° miscut towards a-plane (off-axis to a-plane). The growth process was carried out in an AIXTRON AIX 200 RF LP-MOVPE system using a low temperature AlN nucleation layer. The growth conditions have been varied with respect to the N₂-H₂ carrier gas composition, total flow, V-III ratio, and the growth temperature. The reactor pressure was kept constant at 35 mbar, the lower limit of our system.

The surface morphology was analyzed by using atomic force microscopy (AFM) and scanning electron microscopy (SEM). High-resolution X-ray diffraction (HRXRD) rocking curve measurements for the (0002) reflection were carried out to examine the crystal quality of bulk AlN epitaxial layers. Low-temperature cathodoluminescence provided information about the spectroscopic properties.

Meanwhile, AlN/GaN (4 nm / 4 nm) superlattice structures (21 periods) were grown on 500 nm thick Al₀.₅Ga₀.₅N buffer layers and covered with a 40 nm thick Al₀.₅Ga₀.₅N cap. The growth interruption time was kept 5 s after each layer grown of AlN and GaN. The HRXRD measurement of the ω-2θ scan (0002 reflection) was performed to verify the periodicity and transmission electron microscopy (TEM) was used to examine the abruptness of the interfaces of the superlattice structure.

3. Result and Discussion

It is found that, at the same growth conditions, the full width half maximum (FWHM) value of the X-ray rocking curve for the (0002) reflection of undoped AlN layer grown on c-plane sapphire substrate off-axis to m-plane is comparatively narrower than that on on-axis and off-axis to a-plane. This shows c-plane sapphire substrate off-axis to m-plane is a good choice for the AlN epitaxial growth. However, there is no significant impact on the surface morphology and growth rate of AlN epitaxial layers due to the different orientations of substrates.

Before optimization, the surfaces showed either whisker-like features of rough grainy characteristics, or columnar textures. Moreover, the presence of a large number of hexagonal-shaped pits of varying size, depth and diameter, as shown in fig. 1 (left), was observed. Similarly, the crystalline quality (as measured by the FWHM of HRXRD) of the layers was inferior, too.

When increasing the growth temperature, substantial morphological differences on the surface quality of the bulk AlN layer were observed. AFM measurements exhibited a remarkable reduction of the rms value of the surface roughness from 21.5 nm to 1.5 nm when increasing the growth temperature from 1110°C to 1170°C. However, further increase of growth temperature is restricted by the system limitations. Meantime, it is noticed that the surface roughness decreases with the increase of total flow whereas the FWHM value of the X-ray rocking curve for the (0002) reflection decreases with the decrease of V-III ratio as shown in fig. 2. Moreover, it is observed that the growth rate increases with the decrease of N₂-H₂ ratio as illustrated in fig. 3. This result is somewhat contrary to the observations made by other groups [3] in the high-temperature GaN growth. However, the increased amount of N₂ drastically decreases the substrate temperature [4] which may explain the decreasing growth rate.
Epitaxial Growth of AlN

Fig. 1: AFM image of bulk AlN layer before optimization (left). Many of hexagonal pits are visible on the surface. The rms surface roughness of 2x2 μm scan is 21.5 nm. AFM image of bulk AlN layer after optimization (right). A hexagonal pit formed after the merging of small pits is also shown and the rms surface roughness of 2x2 μm scan is 0.4 nm.

Since the suppression of the pre-reactions between the precursors and the surface diffusion of Al atoms during the growth process are the key factors in determining the surface morphology, the growth process was hence optimized by decreasing the ammonia and trimethylaluminum flow rate and apparently lowering the V-III ratio at higher growth temperature. The flow rate of N₂ and H₂ was increased to keep the total flow constant.

Fig. 2: Change in FWHM value of the X-ray rocking curve for the (0002) reflection with respect to the V-III ratio.

Fig. 3: Change in growth rate with respect to the N₂-H₂ ratio.
As superior CL spectra were ascribed to the samples having lower growth rate, it was customary to keep the \( \text{N}_2\text{-H}_2 \) ratio greater than 1 to maintain the same growth rate. Consequently, the optimized growth process has led to an almost flat surface morphology with a significantly reduced number of hexagonal-shaped pits (approximately \( 5 \cdot 10^7 \text{cm}^{-2} \)) and good crystalline quality. Figure 1 (right) shows the AFM image of a bulk AlN layer after optimization of the growth process. The surface of the AlN layer is a well ordered atomic layer with a measured rms value of the surface roughness of 0.4 nm. The FWHM of the X-ray rocking curve for the (0002) reflection is 200 arcsec. These excellent data were further confirmed by a band edge excitonic emission with a FWHM of 25 meV (fig. 4). The LO phonon replica, shown in the inset of fig. 4, confirm the good optical quality of the bulk AlN epitaxial layers.

![Low-temperature CL spectra of bulk AlN layer. The LO phonon replica are shown in the inset.](image)

**Fig. 4:** Low-temperature CL spectra of bulk AlN layer. The LO phonon replica are shown in the inset.

After having obtained the required crystalline quality and surface morphology of bulk AlN layer, AlN/GaN superlattice structure were grown on c-plane sapphire. Figure 5 (left) illustrates the HRXRD measurement of the \( \omega-2\theta \) scan (0002 reflection) of such a structure which shows the superlattice related satellite peaks (SL) confirming the good periodicity of the layers. The defects and the abruptness of the interfaces of the superlattice structure
Fig. 5: HRXRD measurement of the $\omega$-$2\theta$ scan (0002 reflection) of AlN/GaN superlattice (left). TEM-bright-field image of the superlattice structure (right). The white arrow shows the structure from the cap to buffer layer. Bright and dark layers in superlattice structure are AlN and GaN, respectively were investigated by transmission electron microscopy (TEM). The TEM-bright-field image of the superlattice structure is shown in fig. 5 (right) where the bright layer is AlN and the dark, GaN. Obviously, the interfaces of AlN on GaN are sharper than those of GaN on AlN and many of the threading dislocations coming through the $\text{Al}_{0.5}\text{Ga}_{0.5}\text{N}$ buffer layer are stopped at the interface between the buffer layer and superlattice structure.

4. Conclusion

Undoped AlN layers, with high crystalline quality and almost flat surface morphology, were grown on $0.3^\circ$ miscut towards m-plane oriented c-plane sapphire substrate by LP-MOVPE. After optimization, we achieved an rms value of the surface roughness of 0.4 nm and the FWHM of the X-ray rocking curve for the (0002) reflection of 200 arcsec. The low-temperature CL spectra demonstrates the good optical quality of the AlN layer. Consequently, it was possible to grow AlN/GaN superlattice structures having good periodicity and abruptness of the AlN/GaN interfaces.

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References


