

## Growth of Single-Crystal (0001) GaN Films on (0001) Sapphire Substrates Using h-BN Buffer Layers by Molecular Beam Epitaxy

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### Abstract

Gallium nitride films were grown on atomically flat and ultrathin h-BN buffer layers on (0001) sapphire substrates using plasma-assisted molecular beam epitaxy. X-ray diffraction revealed that single-crystal (0001) GaN films were grown on the 1.5- and 3-nm-thick h-BN buffer layers. The intensity of (0002) GaN in X-ray diffraction decreases as the thickness of the h-BN buffer layers increases, indicating that a h-BN buffer layer with a thickness of less than 1 nm or an AlN layer on the h-BN buffer is necessary to improve the crystalline quality of the GaN.

### 1. Introduction

Hexagonal boron nitride (h-BN) has a graphite-like honeycomb structure. Previous research has demonstrated that a h-BN layer inserted between a sapphire substrate and GaN-based device structure grown on the sapphire substrate acts as both a buffer layer for the growth of a high-quality GaN-based device structure and as a release layer that enables the mechanical transfer of the GaN-based device structure onto a foreign substrate [1]. It was also found that an AlN or AlGa<sub>N</sub> layer on the h-BN buffer layer makes it possible to grow a single-crystal GaN film on the AlN or AlGa<sub>N</sub> on the h-BN layer [1]. However, GaN grown directly on the ultrathin h-BN layer with metalorganic vapor phase epitaxy (MOVPE) had a rough and irregular island-shaped surface morphology and was polycrystalline.

Hexagonal boron nitride epitaxial growth has been achieved with MOVPE [1]. During BN MOVPE, a strong parasitic reaction occurs between triethylboron (TEB) and ammonia (NH<sub>3</sub>) [2]. The thickness of the h-BN buffer layer has a large effect on the crystalline nature of the GaN film grown on the buffer layer. However, the strong parasitic reaction makes it difficult to control the thickness of the h-BN at the atomic scale. In contrast to MOVPE, the strong parasitic reactions between TEB and NH<sub>3</sub> can be avoided in molecular beam epitaxy (MBE). However, until now, very little research has been conducted on MBE growth of h-BN [3]. Here, we report on the growth of (0001) single-crystal GaN thin films on 1.5- and 3-nm-thick h-BN buffer layers on (0001) sapphire substrates using MBE.

### 2. Experiments

The h-BN buffer layers and GaN thin films were grown on single-crystal double-side polished (0001) sapphire substrates with plasma-assisted MBE. The growth temperatures of the h-BN buffer layers and GaN thin films were 1000 °C and 800 °C, respectively, measured with a thermocouple. Reactive nitrogen was supplied by flowing high-purity N<sub>2</sub> gas (99.99995%) through the RF plasma source operating at 400 W with an N<sub>2</sub> flow rate of 1.6 sccm. High-purity B (99.9999%) was evaporated with an electron-beam gun, and metallic Ga was supplied with a standard effusion cell as the group-III sources. Three sample types were fabricated. As a reference, 300-nm-thick GaN film was grown directly on the sapphire substrate with nitridation of a (0001) sapphire substrate. Using the same growth conditions for the GaN film as the reference film, we grew 300-nm-thick GaN films on 1.5- and 3-nm-thick ultrathin h-BN buffer layers on (0001) sapphire substrates. The thickness of the h-BN layer was controlled by an Inficon XTC/2 thin-film-deposition controller.

The thicknesses of the h-BN buffer layers and the crystalline qualities of the GaN films were characterized with X-ray reflectance and X-ray diffraction (XRD) in a 2θ/ω configuration, respectively, using an X-ray diffractometer (Rigaku SmartLab) with a wavelength of 1.541 Å for a copper target. The surface morphologies of the h-BN buffer layers and the GaN films were analyzed with atomic force microscopy (AFM) using an SII NanoTechnology NanoNav E-sweep.

### 3. Results and discussions

Figure 1 shows the X-ray reflectance curve for the h-BN layer grown with MBE. The reflectivity clearly oscillated two times, indicating that the surface of the h-BN layer was relatively flat. Figure 1 also plots a simulated X-ray reflectance curve obtained by fitting to the measured reflectance using a layered model. The fitting curve is in good agreement with the measured one, and the thickness of the h-BN layer was estimated to 3.0 nm. The root mean square (RMS) roughness of the 3-nm-thick h-BN layer was found to be 0.16 nm by AFM. Reflection high-energy electron diffraction for the 3-nm-thick h-BN revealed a streaked (1 × 1) pattern, indicative of an atomically flat h-BN surface [4].

X-ray diffraction for the GaN film grown directly on the

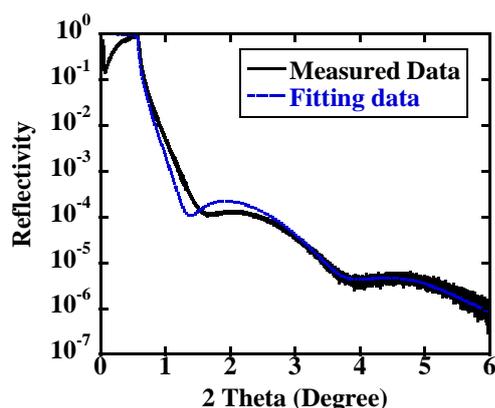


Fig. 1 X-ray reflectance curves of 3-nm-thick h-BN layer grown with MBE. Solid and dotted curves correspond to measured X-ray reflectivity and simulated reflectivity, respectively.

sapphire substrate exhibits only a GaN (0002) diffraction peak, indicating that a (0001) single-crystal GaN film was grown on the (0001) sapphire substrate. The AFM image for the GaN film shows a flat GaN surface.

Figure 2 shows X-ray diffraction for the GaN film grown on a 1.5-nm-thick h-BN buffer layer. Only a GaN (0002) diffraction peak was observed, indicating a (0001) single-crystal GaN film was grown on the 1.5-nm-thick h-BN buffer layer. The GaN film grown on the h-BN buffer layer with MOVPE was polycrystalline [1]. The difference between the GaN growth temperature for MOVPE (1000 °C) and for MBE (800 °C) resulted in the crystalline difference between the MOVPE-grown and MBE-grown GaN films. However, the intensity of the GaN (0002) peak for the GaN film grown on 1.5-nm-thick h-BN decreases drastically compared with that of the GaN film grown directly on the sapphire substrate. X-ray diffraction for the GaN film grown on the 3-nm-thick h-BN buffer layer also shows only a GaN (0002) diffraction peak. The intensity of the GaN (0002) peak for the GaN film grown on 3-nm-thick h-BN decreases compared with that of the GaN film grown on the 1.5-nm-thick h-BN.

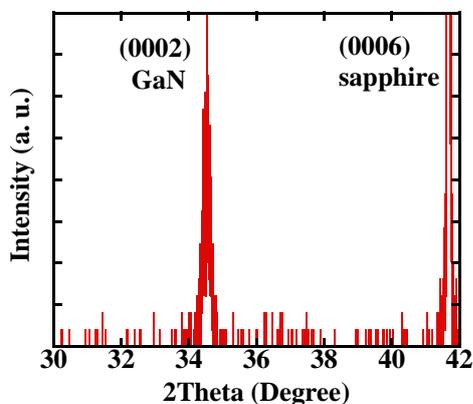


Fig. 2 X-ray diffraction using  $2\theta/\omega$  configuration for GaN film grown on 1.5-nm-thick h-BN buffer layer on (0001) sapphire substrate.

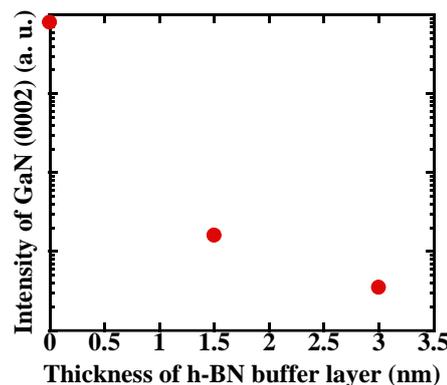


Fig. 3 Intensity of GaN (0002) diffraction peak in XRD using  $2\theta/\omega$  configuration as a function of thickness of h-BN buffer layer.

Figure 3 shows the intensity of the GaN (0002) diffraction peak in X-ray diffraction as a function of the thickness of the h-BN buffer layer. Compared to the peak intensity for the GaN film grown directly on the (0001) sapphire substrate, the peak intensity decreases drastically as the thickness of the h-BN buffer layer increases. AFM images for the GaN films grown on the 1.5- and 3-nm-thick h-BN buffer layers show a three-dimensional rough surface. These results indicate that thicker h-BN buffer layers deteriorate the crystalline quality of the GaN films. Thinner h-BN buffer layers with a thickness of less than 1 nm make it possible to improve the surface morphology and crystalline quality of the GaN films. In addition, during MOVPE growth, a single-crystal (0001) GaN film was grown on an AlN or AlGaIn layer on the h-BN buffer layer [1]. Therefore, during MBE growth, an AlN or AlGaIn layer grown on the h-BN buffer layer may also enable the improvement of the crystalline quality of the GaN film.

#### 4. Conclusions

We grew single-crystal (0001) GaN films on 1.5- and 3-nm-thick h-BN buffer layers on (0001) sapphire substrates. The intensity of the GaN (0002) diffraction peak in X-ray diffraction decreases, and the surface morphology becomes rough, as the thickness of the h-BN buffer layer increases.

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