Real-Time Measurements of Wing Tilt during Lateral Epitaxial Overgrowth of GaN

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Introduction

We have performed *in situ*, real-time x-ray diffraction measurements to study the lateral epitaxial overgrowth (LEO) of GaN. The LEO technique consists of coating a heteroepitaxial film with a patterned mask and performing subsequent additional growth such that the material growing through the mask openings continues its growth laterally over the mask. This overgrowth process in GaN results in a reduction in threading dislocation density in the overgrowth region compared to that obtainable in the initial template film. This improved material has directly led to improved performance in GaN-based optoelectronic devices, such as blue lasers, light-emitting diodes, and p-n junctions with lowered leakage currents.

Although the improved quality of the LEO growth is important in improving device performance, difficulties remain in controlling the structural quality of the overgrowth material. In particular, the crystal planes in the "wings" (overgrowth region) are tilted with respect to those in the "window" (seed area) regions. Therefore, the boundary of overgrowth wings from neighboring stripes forms a tilt boundary. By performing *in situ*, real-time x-ray diffraction measurements in the organic metallic vapor phase epitaxy environment (OMVPE), we have directly observed the emergence and evolution of wing tilt that occurs during the lateral overgrowth of GaN on stripes patterned in a SiO₂ masks.¹

Methods and Materials

Templates for the overgrowth experiments were prepared at University of California at Santa Barbara. Epitaxial 2-3 μ m GaN films were grown by metal-organic chemical vapor deposition on sapphire. The GaN was coated with a 200 nm mask of SiO₂. Openings in the SiO₂ were patterned as stripes with 5 μ m opening width and 20 μ m period. These opening stripes were oriented along the [1 0 1 0] direction in the GaN.

The *in situ* x-ray diffraction experiments on LEO growth were performed at the Basic Energy Sciences Synchrotron Radiation Center (BESSRC) undulator beamline 12-ID-D at the Advanced Photon Source (APS). This beamline has a dedicated vertical two-flow metal-organic chemical vapor deposition chamber mounted on a z-axis surface diffractometer.^{2,3} A relatively high x-ray energy of 24 keV (0.517 Å) was used to penetrate the 2-mm-thick quartz walls of the chamber. LEO growth was performed at 1060°C using trimethylgallium (TMGa) and ammonia (NH₃) precursors. Total chamber pressure was 76 Torr. The H₂/N₂ ratio was 1:3. During growth, the TMGa and NH₃ flows were 4.56 μ mol/min and 1.04 slpm, respectively; the conditions were optimized with respect to obtaining LEO stripes with a rectangular cross section.

Results

Figure 1 shows the x-ray scattering data of the development of the tilt during a 3600 s duration LEO growth at 1060°C. The central peak is the 1 0 $\overline{1}$ 3 Bragg peak from the underlying bulk



FIG. 1. Evolution of the intensity of the GaN 1 $0\overline{1}$ 3 peak during OMVPE growth of the GaN shows the evolution of the tilt in the LEO wing material with respect to the initial template.

GaN. The tilting of the crystal planes in the overgrowth with respect to the underlying template was monitored by measuring the splitting of the peak in the direction of the tilt. The emergence of features distinct from the central peak is evident for times as early as 100 s. By 300 s into the growth, distinct wing peaks have evolved, corresponding to a tilt of the overgrowth of $\sim 0.9^{\circ}$. As growth continued, the wings narrowed, and gradually increased in separation from the central peak, reaching a tilt of ~1.19° after 3600 s of growth. Growth was halted at this point and the sample cooled to room temperature. Upon cooling, the tilt of the overgrowth did not change significantly, increasing from 1.19° at 1060°C to 1.36° at room temperature. Scanning electron microscopy characterization of post-growth morphologies was performed by examining a time sequence of samples with identical growth conditions. We note that these microscopy studies showed that, at 200 s, the LEO GaN filled the mask window opening, and, at 400 s, overgrowth over the mask had already begun.

Discussion

Our studies show that the wing tilt develops rapidly during the initial growth of the overgrowth. Although the origins of the overgrowth tilt are unknown, this work shows that thermally induced stresses during cooling appear to have little effect on tilt, contrary to what may be inferred from recent modeling studies.⁴ The tilt is well developed during growth, implying that other factors are critical, such as stress states early in growth and changes in the physiochemical properties of the mask.

Acknowledgments

This work made use of the UCSB MRL Central Facilities supported by the National Science Foundation (Grant No. DMR-9632716). This work was supported by the Office of Naval Research (C. Wood), the National Science Foundation (Grant No. DMR-9704201), the State of Illinois (HECA), and the U.S. Department of Energy, BES under contract W-31-109-ENG-38. **References**

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