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# Study of the Epitaxial Lateral Overgrowth (ELO) Process for GaN on Sapphire Using Scanning Electron Microscopy and Monochromatic Cathodoluminescence

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Growth of GaN by MOVPE on mismatched substrates such as sapphire or SiC produces a columnar material consisting of many hexagonal grains ~1 µm across. In contrast, the epitaxial-lateral-overgrowth (ELO) process creates a new material — single-crystal GaN. We have studied the ELO process using GaN/sapphire layers patterned with $SiO_2$ stripes. SEM images show that the (0001) GaN surface remains very flat as the ELO progresses. Cathodoluminescence images at 590 nm reveal spotty yellow-green emission from the columnar GaN as it emerges from the window areas. Very bright 590 nm emission occurs as the ELO process begins. We associate this deep-level cathodoluminescence with the strain field that accompanies the conversion of columnar GaN into single-crystal GaN via the ELO process. As the ELO process continues across the $SiO_2$ stripes, the 590 nm emission disappears and is replaced with pure band edge cathodoluminescence at 365 nm which is maintained until coalescence of adjacent ELO layers occurs near the centers of the $SiO_2$ stripes.

## 1 Introduction

The development of III-V nitride materials and devices has suffered from the lack of availability of low-dislocation-density, lattice-matched native nitride substrates for device synthesis by MOVPE or MBE. As a consequence, growth of GaN and other III-V nitrides on mismatched substrates such as sapphire or SiC produces a columnar material consisting of many small hexagonal grains [1]. The individual grains have a distribution of tilt and rotation within the GaN film, as is illustrated in Figure 1, which gives rise to very large dislocation densities. In spite of this, there have been demonstrations of very bright LEDs and laser diodes at Nichia Chemical [2] [3] [4] [5] and elsewhere [6] [7] [8] [9]. These light-emitting devices, all prepared by MOVPE, show dislocation densities of $10^9$-$10^{10}$ per $cm^2$ but function as bright light emitters as though these internal disruptions to periodicity are virtually absent – perhaps due to some unknown passivation process associated with the MOVPE growth process itself.

Recently, however, there have been demonstrations of defect reduction in GaN layers grown on sapphire [10] [11] [12] [13] and SiC [14] [15] [16] using an epitaxial lateral overgrowth (ELO) technique. The ELO technique is illustrated schematically in Figure 2. The ELO process produces stripes of GaN (5–10 µm wide) with a remarkable reduction in dislocation density to about $10^4$ per $cm^2$ or less. It is on these low-dislocation-density single-crystal GaN stripes that Nichia Chemical has produced laser diodes having very long CW lifetimes (10,000 hrs) [17].

In this paper, the results of a systematic study of GaN ELO on sapphire are reported. Sequential scanning electron microscope (SEM) photomicrographs were obtained for a series of GaN ELO samples which provide images of the ELO growth process from its initiation to its completion (complete coalescence of GaN at the centers of the $SiO_2$ stripes). In addition, monochromatic cathodoluminescence images of the GaN ELO samples were obtained which clearly show the spatial distribution of band edge CL (365 nm) versus deep level defect-band CL (~590 nm) for each GaN ELO sample.



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## 2 Experimental Details

The GaN ELO samples were prepared in an MOVPE system that is part of a five-chamber UHV-interconnected nitride "cluster tool" at NCSU consisting of

1. Hydrogen Plasma chamber for wafer surface cleaning,

2. Auger Spectroscopy chamber for surface chemistry studies,

3. Molecular Beam Epitaxy system equipped with an ammonia source, two plasma nitrogen sources, a plasma hydrogen source, five MBE effusions cells, and *in situ* RHEED and optical pyrometry,

4. Bulk Nitride Crystal Growth system which employs modified Sublimation Vapor Phase Epitaxy (SVPE), and

5. Metallorganic Vapor Phase Epitaxy (MOVPE) system for the synthesis of nitride thin films and devices.

Each chamber is equipped with a load-lock so that substrate wafers up to 75 mm in diameter may be transferred into and out of all of the chambers under UHV conditions.

Two views of the MOVPE system in which the GaN ELO experiments were performed are shown in Figure 3 and Figure 4. The MOVPE system was designed and built at NCSU. It features vertical gas flows and fast substrate rotation (up to 2000 rpm). In addition, the MOVPE system is equipped with a substrate mount and heater assembly of original design which can be used to obtain substrate temperatures up to 1200 °C, as measured by optical pyrometry. In the ELO experiments described herein, 50 mm diameter sapphire wafers were employed as base substrates for ELO of GaN. ELO of GaN was performed in the NCSU MOVPE chamber using trimethylgallium (TMGa) and ammonia ($NH_3$) precursors. Hydrogen was employed as the carrier gas at a reactor pressure of 76 Torr. Growth temperatures were 1060-1080 °C and ammonia flows were varied between 0.15 and 0.25 mole/min. TMGa flows of 20-40 µmole/min were used which resulted in very flat 2D planar growth of GaN at rates of 1-2 µm/hr.

For GaN ELO on sapphire, the first step was to grow a conventional low-temperature (~500 °C) buffer layer of GaN on (0001) sapphire followed by the deposition of 1-2 µm of GaN at high temperatures (~1050 °C) by MOVPE. This produced a GaN surface containing ~$10^9$-$10^{10}$ dislocations per $cm^2$ [1]. The next step was to cover the entire wafer with ~100 nm of $SiO_2$. Windows in the $SiO_2$ were then obtained by conventional photolithography and $SiO_2$ etching to expose parallel stripes of GaN separated by stripes of $SiO_2$. These stripes were oriented along a $[1\,\bar{1}\,0\,0]$ GaN crystal direction. Because of the 30 degree rotation of GaN epitaxy on basal-plane sapphire, the GaN stripe length corresponds to the direction of a line drawn from the center of the sapphire wafer that is perpendicular to the $[1\,1\,\bar{2}\,0]$ sapphire flat. Three window/stripe widths of 3 µm/4 µm, 3 µm/5 µm, and 4 µm/8 µm, respectively, were employed in the ELO experiments (see Figure 5).

The GaN ELO samples were characterized using a JEOL JSM-6400 scanning electron microscope (SEM) equipped with a model SP13064-6400 digital scan generator (DSG). Digital electron microscope images are acquired by the DSG with the aid of a computer by taking control of the electron beam positioning and digitizing the video signal output of the instrument. The DSG controls the instrument's horizontal and vertical scan coils, acquires a video signal at each pixel, and forms a digital image. Image pixel densities of up to 2560 x 1920 pixels are then stored on computer as a high-resolution digital image. In the present study, the JEOL DSG was employed to produce digital SEM images of the GaN ELO process from start to finish that have remarkable clarity and resolution.

The SEM is also equipped with an Oxford Instruments Mono-CL cathodoluminescence (CL) accessory. With this unit it is possible to obtain spectral CL scans from 180 nm to 900 nm. In addition, monochromatic CL images of samples under investigation can be obtained at any selected wavelength in this wavelength range. In the present study, digital CL images at 590 nm (deep level emission) and at 365 nm (band edge emission) were obtained for selected GaN ELO samples. These images show the CL as a white emission on a dark non-emitting background. However, since the wavelength of each monochromatic CL image was known, each image was colored using Adobe Photoshop 4.0 for display purposes. The 590 nm emission was colored yellow-green and the 365 nm emission was colored violet so that both deep level and band edge emissions from the GaN ELO samples are visible to the human eye.

## 3 Results and Discussion

Growth of GaN by MOVPE under the appropriate deposition parameters (substrate temperature, III-V ratio, carrier gas flow geometry, etc.) promotes an epitaxial lateral overgrowth of GaN onto the $SiO_2$ stripes after the initial growth of GaN, occurring only on the window areas, reaches the level of the $SiO_2$ surface (~100nm). During this initial GaN growth sequence, the $Si0_2$ stripes function as non-wettable surfaces and no GaN deposition occurs on them. However, once the GaN film growth from the window stripes reaches the tops of the $SiO_2$ stripes, epitaxial lateral overgrowth of GaN commences.

Sequential SEM images of the GaN ELO process are shown in Figures 6 - 11, respectively. Figure 6 shows the beginning of the ELO growth process for GaN with lateral sidewalls ({1 $\bar{1}$ 0 1} planes) spreading onto the $SiO_2$ stripes. GaN growth over the window areas is two-dimensional (2D) and very flat. Figure 7, Figure 8, and Figure 9 show the ELO growth of GaN spreading increasingly onto the $SiO_2$ stripes while maintaining a 2D surface. Figure 10 shows a view of the GaN ELO process when coalescence of adjacent GaN layers near the centers of the $SiO_2$ stripes is ~95% complete. The GaN surface remains 2D and very flat. Figure 11 shows the GaN ELO after complete coalescence. The GaN surface remains flat over most of the ELO areas. Note, however, the texture that is present near the centers of the $SiO_2$ stripes where the coalescence between adjacent single-crystal GaN stripes occurs. We believe that this texture is real and occurs because adjacent GaN ELO single-crystal stripes are not in registry with one another. As a consequence, the flat ELO growth is modified near the centers of the $SiO_2$ stripes as adjacent GaN single-crystal stripes "collide" with one another, a process that has also been shown to generate a line of dislocations near the centers of the $SiO_2$ stripes [17].

Monochromatic CL images of the ELO process are shown in Figures 12 and 13. In Figure 12, a CL image at 590 nm of deep-level defect band emission from a GaN ELO sample is shown. It is seen from the figure that deep-level emission is present from the window areas as a non-uniform speckled emission. This is the emission from the individual grains of the columnar GaN that grows over the window areas. Note that emission from individual grains is resolved, but not all of the grains over the window areas emit CL. In contrast to this, very bright and uniform 590 nm emission is seen from the initial ELO overgrowth regions where columnar GaN is converted into single-crystal GaN. The conversion of columnar GaN into single-crystal GaN must be accompanied by a large strain field, since the original GaN columns are not in single-crystal registry. We believe that the observed uniform emission (no grains) from these initial ELO areas is the CL signature of strained single-crystal GaN. Note that deep-level emission at 590 nm is completely absent after ELO growth of ~1 µm toward the centers of the $SiO_2$ stripes. This region of the ELO process produces strain-free single-crystal GaN.

In Figure 13, a CL image at 365 nm of band-edge emission from a GaN ELO sample is shown. This UV emission has been colored violet as an aid to the eye. The band edge CL emission is very weak from the window areas where the GaN is columnar. By comparing Figure 12 with Figure 13, it is seen that mixed 590 nm and 365 nm CL emission occurs from the initial ELO overgrowth regions where columnar GaN is converted into single-crystal GaN. Intense band edge emission at 365 nm, with the deep-level 590 nm emission absent, occurs over most of the remainder of the ELO areas on $SiO_2$. This is the CL signature of unstrained single-crystal GaN. Note, from the figure, that the ELO process is not quite completed at the centers of the $SiO_2$ stripes for this sample.

Results similar to those reported above were obtained using 3 µm x 4 µm and 4 µm x 8 µm ELO samples like those shown in Figure 4. The initial ELO on these samples clearly showed the conversion of columnar GaN into single-crystal GaN with an accompanying strain field. As the ELO process continued towards coalescence, only band-edge emission was observed from these strain-free single-crystal regions.

The MOVPE growth parameters employed during the present study produced a lateral growth to vertical growth ratio of about 1:1. We are presently exploring new MOVPE growth parameters which, hopefully, will increase the lateral growth rate relative to the vertical growth rate so that much wider single-crystal stripes of GaN may be obtained via the ELO process.

## 4 Summary

The ELO process for GaN/sapphire has been studied by SEM and monochromatic CL. The SEM images show how the flat GaN ELO process spreads onto the $SiO_2$ stripes with {1 $\bar{1}$ 0 1} growth front sidewalls. Remarkable CL images were obtained which show the conversion of columnar GaN into single-crystal GaN at the initiation of ELO process with an accompanying strain field. Beyond this strain field, the CL from the ELO regions consists of pure band edge emission at 365 nm.

## ACKNOWLEDGMENTS

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## FIGURES

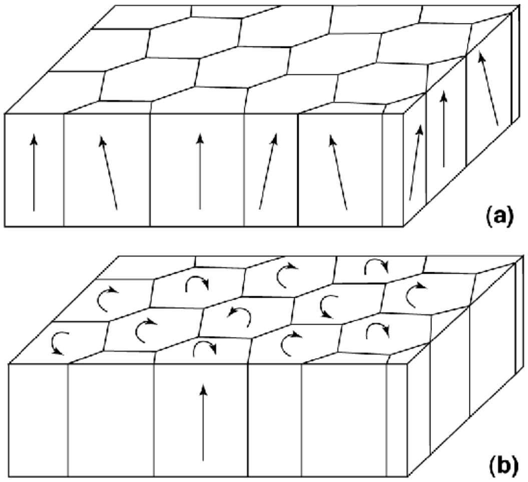


Figure 1. Columnar model for the III-V nitrides [1]. Nitride films prepared by MOVPE consist of a mosaic of vertical columns or grains ~1 µm across. There is a slight distribution in intergrain orientation consisting of two components: (a) tilt (~5 arc min) and (b) twist (~8 arc min).

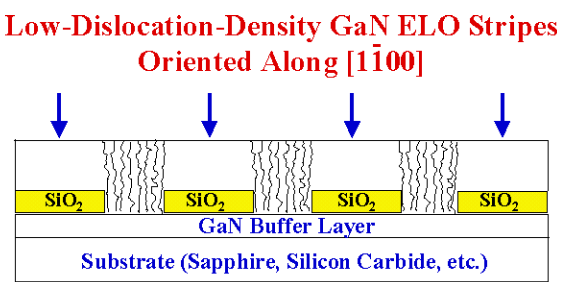


Figure 2. Schematic of ELO process for GaN. The GaN emerging from the window regions has a high density of threading dislocations (~$10^{10}$ per cm$^2$), whereas the GaN overgrowth on the Si0$_2$ stripes has a drastic reduction in dislocation density to ~$10^4$ per cm$^2$ or less.





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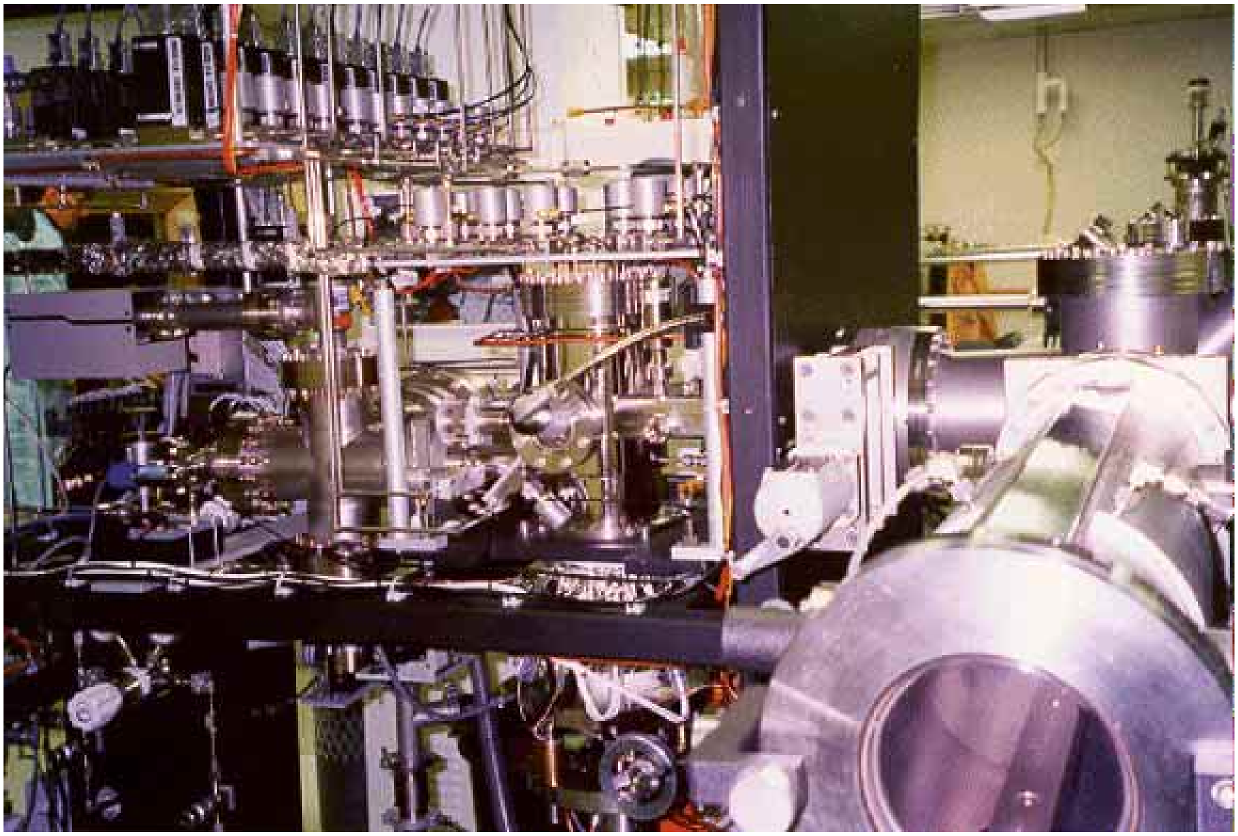

Figure 3. Photograph of MOVPE system showing UHV load-lock and linkage to the five-chamber nitride "cluster tool" at NCSU

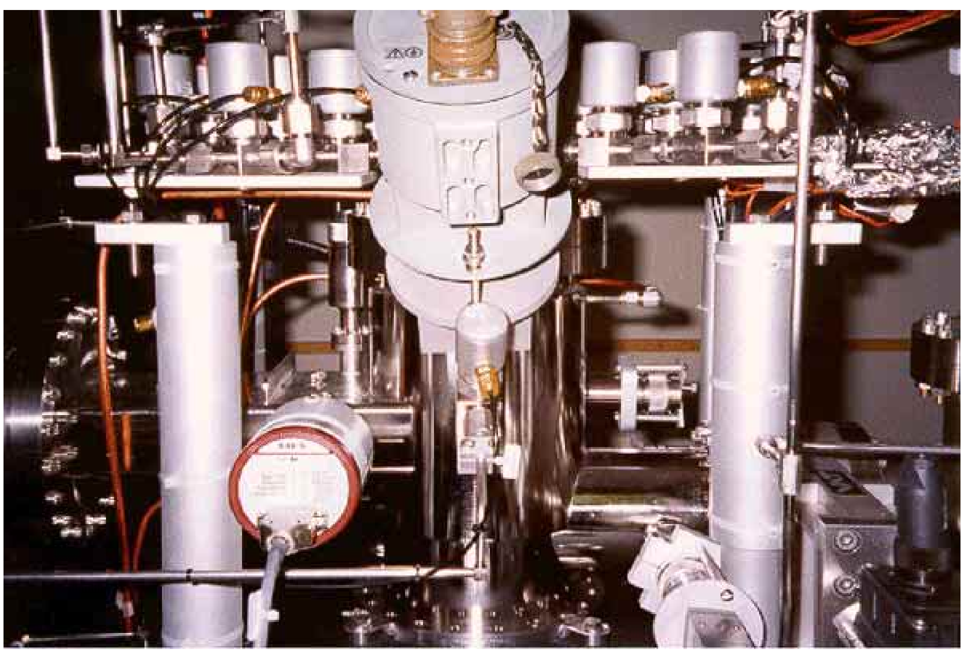

Figure 4. Close-up photo of MOVPE system which features vertical gas flows and fast substrate rotation (to 2,000 rpm). The MOVPE system employs a special substrate heater assembly for substrate heating up to 1200 °C and an optical pyrometer for substrate temperature determinations. The entire MOVPE film growth sequence is controlled by computer.




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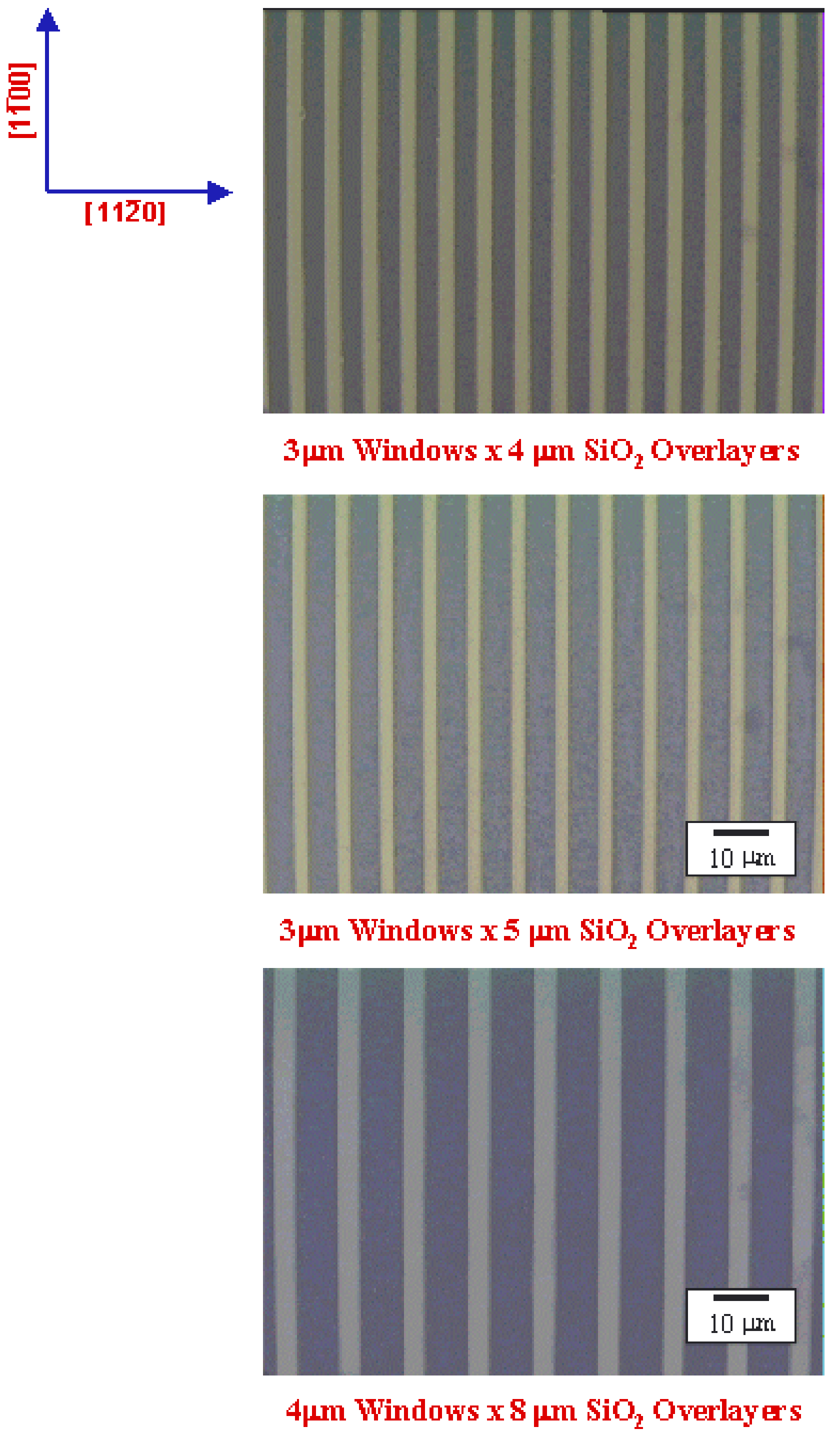


Figure 5. Nomarski interference-contrast micrographs of GaN/sapphire samples processed for use in GaN ELO experiments.




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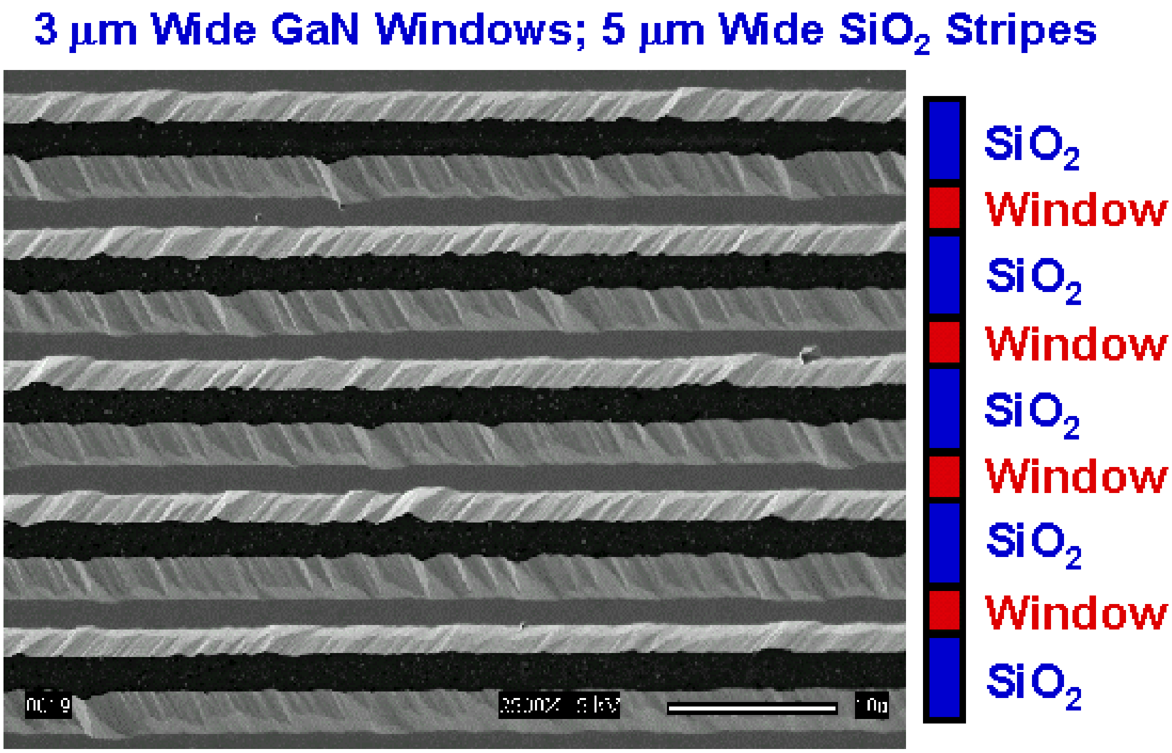


Figure 6. ELO growth of GaN begins with lateral sidewalls visible. The sidewalls correspond to $\{1\,\bar{1}\,0\,1\}$ planes. GaN growth over window areas is very flat two-dimensional (2D) growth. The bare $SiO_2$ stripes are the black areas in the figure.

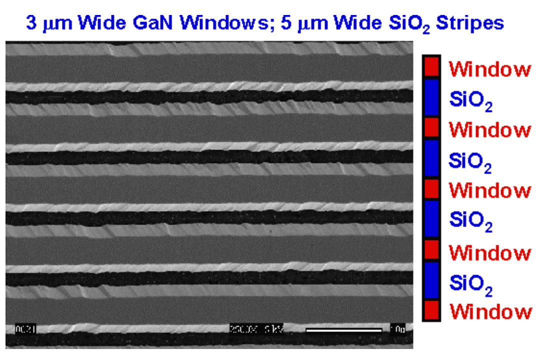


Figure 7. ELO growth continues with lateral sidewalls visible. 2D growth of GaN spreads laterally onto $SiO_2$ stripes.

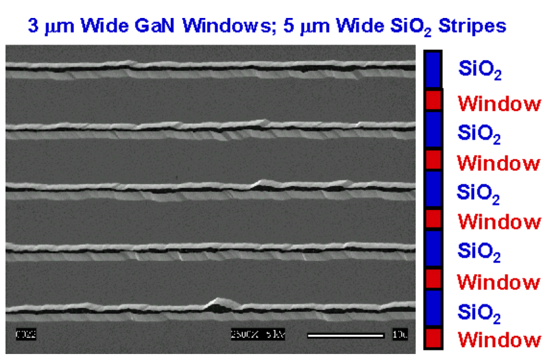


Figure 8. ELO growth process continues with flat 2D growth of GaN continuing to spread laterally onto the $SiO_2$ stripes.

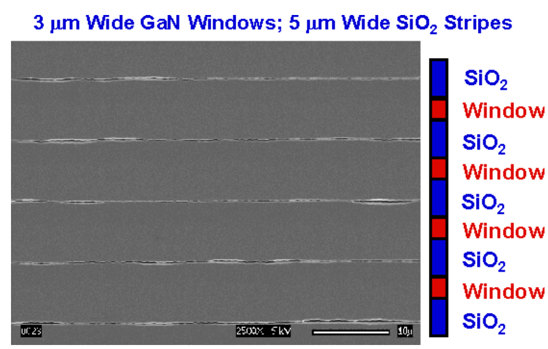


Figure 9. ELO growth process continues with flat 2D growth of GaN maintained. Adjacent GaN ELO layers are beginning to coalesce near the centers of the $SiO_2$ stripes.

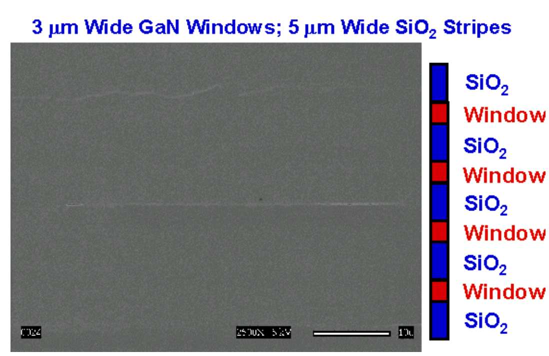


Figure 10. Coalescence of GaN ELO layers is ~95% complete. GaN surface remains very flat.




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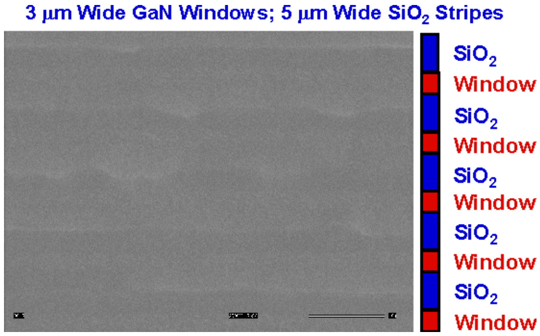


Figure 11. Coalescence of GaN ELO layers is ~100% complete. GaN surface remains very flat. Note, however, the texture that is present near the centers of the $SiO_2$ stripes where the coalescence between adjacent single-crystal GaN stripes occurs.

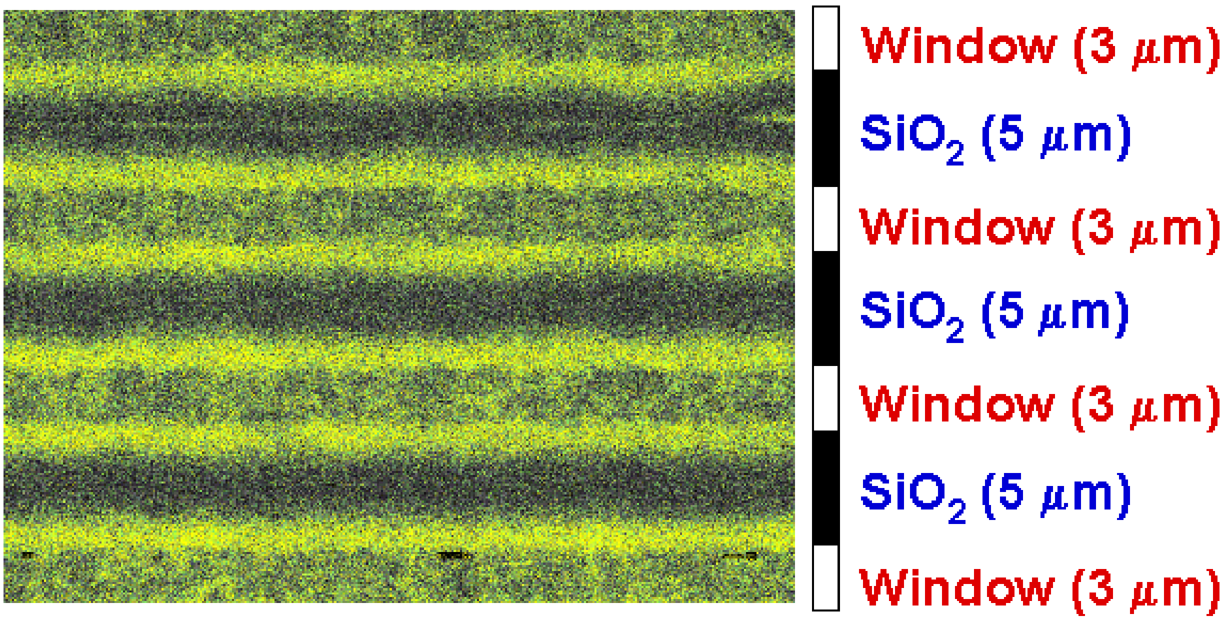


Figure 12. Cathodoluminescence image at 590 nm of deep level defect band emission from GaN. Deep level emission is present for window areas and for initial ELO overgrowth regions where columnar GaN is converted into single-crystal GaN with an accompanying large strain field. Deep level emission is completely absent over the remaining ELO areas toward the centers of $SiO_2$ stripes. This is strain-free single-crystal GaN.



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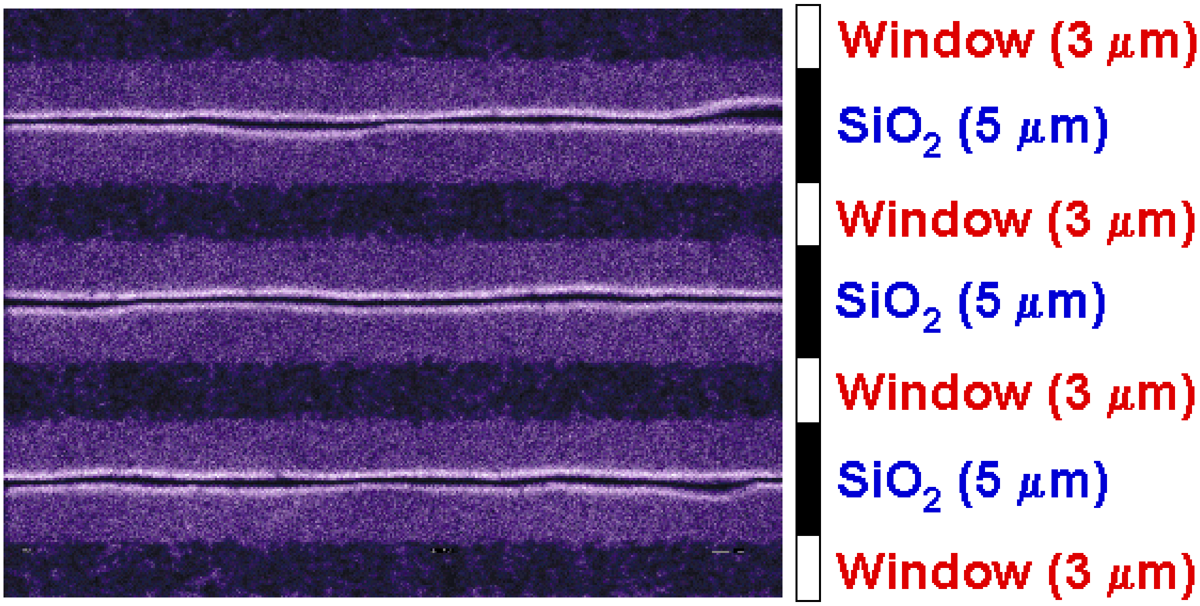


Figure 13. Cathodoluminescence image at 365 nm of band edge emission from GaN ELO sample (colored violet). The band edge emission is very weak from window areas where GaN is columnar. Mixed 365 nm and 590 nm emission occurs from the initial ELO overgrowth regions near windows where columnar GaN is converted into single-crystal GaN. Intense band edge emission, with the 590 nm emission absent, occurs over most of the remainder of the ELO areas on $SiO_2$. This is unstrained single-crystal GaN. The ELO process is not quite completed at the centers of $SiO_2$ stripes for this sample.



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