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Final LDRD report: Low Dislocation GaN via Defect-Filtering, Self-Assembled SiO₂-Sphere Layers

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Low Dislocation GaN via Defect-Filtering, Self-Assembled SiO₂-Sphere Layers

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Abstract

The III-nitride (AlGaInN) materials system forms the foundation for white solid-state lighting, the adoption of which could significantly reduce U.S. energy needs. While the growth of GaN-based devices relies on heteroepitaxy on foreign substrates, the heteroepitaxial layers possess a high density of dislocations due to poor lattice and thermal expansion match. These high dislocation densities have been correlated with reduced internal quantum efficiency and lifetimes for GaN-based LEDs. Here, we demonstrate an inexpensive method for dislocation reduction in GaN grown on sapphire and silicon substrates. This technique, which requires no lithographic patterning, GaN is selectively grown through self-assembled layers of silica microspheres which act to filter out dislocations. Using this method, the threading dislocation density for GaN on sapphire was reduced from $3.3 \times 10^9 \text{ cm}^{-2}$ to $4.0 \times 10^7 \text{ cm}^{-2}$, and from the 10^{10} cm^{-2} range to $\sim 6.0 \times 10^7 \text{ cm}^{-2}$ for GaN on Si(111). This large reduction in dislocation density is attributed to a dislocation blocking and bending by the unique interface between GaN and silica microspheres.

Acknowledgments

We thank Jeffrey Figiel for technical assistance with GaN epilayer growth, and Daniel D. Koleske for exploratory InGaN quantum well growth on one of our samples. We acknowledge support from Sandia's Laboratory Directed Research and Development program. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract No. DE-AC04-94AL85000.

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1. Introduction

The purpose of this project is to develop a novel and inexpensive technique for the growth of low-dislocation density GaN using self-assembled silica sphere layers as a dislocation “blocking” mask. The III-nitride (AlGaInN) materials system forms the foundation for white solid-state lighting (SSL), the adoption of which could result in very significant energy savings. Due to the lack of availability, small size, and unfavorable economics of bulk GaN substrates, the growth of GaN-based devices, including blue-green-yellow LEDs for SSL, has relied on heteroepitaxy on foreign substrates, most commonly sapphire. However, the large lattice mismatch (~14%) of sapphire with GaN results in high threading dislocation densities (TDDs) as high as $\sim 10^{10} \text{ cm}^{-2}$ in GaN films grown directly on sapphire.[1] At increasingly high power densities, device lifetimes become dramatically shortened due to the presence of high dislocation densities, and there is also substantial evidence that thermal conductivity and internal quantum efficiency (IQE) decreases as dislocation density increases.[2]

In order to achieve DOE’s targeted efficiency and device lifetime goals for SSL, it will thus be necessary to provide low dislocation density GaN substrates that will allow fabrication of high power density, efficient LEDs with long lifetimes. Additionally, the costs of these substrates and buffers must be low enough to allow LEDs to be cost-competitive with other lighting technologies, including fluorescent lighting. Epitaxial lateral overgrowth (ELO) techniques are currently employed to further reduce the dislocation densities of heteroepitaxial GaN down to the mid-high 10^7 cm^{-2} range.[3] However, because of the process complexity of ELO, which requires a simple buffer, followed by alternating lithographic masking and growth steps for each level of defect blocking, the epitaxial cost is roughly tripled or more. This cost is prohibitive for inexpensive, high power LEDs and hence ELO is primarily used only for the fabrication of more costly devices such as blue laser diodes. While the size, cost, and availability of high quality bulk freestanding GaN substrates are likely to continue to improve in the future, the cost is unlikely to come close to that of sapphire substrates for the foreseeable future, making them impractical for inexpensive devices like LEDs.

2. Technical Approach

Our technical approach for the growth of low-dislocation density GaN takes advantage of self-assembly and selective area growth principles. This technique differs fundamentally from current ELO schemes by employing a self-assembled, multi-layered growth template of close-packed SiO₂ spheres to filter vertical dislocations. This “bottom-up” approach requires no lithographic patterning and allows for a single GaN growth step through one or more levels of defect filtering sphere layers, without growth interruption or additional lithography for each level of defect filtering. The number of defect filtering levels is controlled simply by how many sphere layers are initially deposited onto the substrate prior to growth. Hence, four levels of defect blocking would require nominally the same cost as a single level of defect blocking, whereas in current ELO techniques the cost increases dramatically for each additional level of defect blocking (e.g. double and triple ELO). This innovative approach could thus allow for the growth of very high quality GaN at a cost comparable to standard GaN heteroepitaxy on sapphire.

The technique is described in further detail as follows. First, monodisperse, nano-to-micrometer sized SiO₂ spheres are self-assembled on a desired growth substrate to form multiple hexagonal close-packed layers with an interconnected network of voids. Due to the 3D hexagonal close packing where spheres and voids alternate positions in adjacent layers, each successive layer of spheres will serve to block and filter out the vertically propagating dislocations, which will terminate upon hitting a sphere. Because dislocations from heteroepitaxial GaN growth typically propagate vertically, they will be blocked by each subsequent layer of spheres and are thus prevented from propagating into the next layer. Each additional sphere layer beyond the first, the number of which can be controlled during the spin-on process, serves as a defect blocking filter. Hence, a three-layer sphere assembly would provide two levels of defect blocking. Figure 1 shows self-assembled, close-packed layers of 1 μm SiO₂ spheres on a sapphire substrate using a modified dip-coating method.

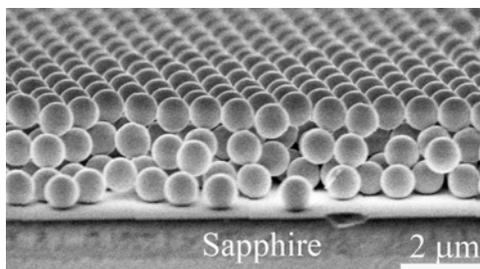


Figure 1. SEM image showing deposition of three layers of hexagonal close-packed 1- μm SiO_2 spheres on sapphire

Following deposition of the desired number of sphere layers onto the substrate, selective growth of GaN on the substrate and through the void network inside the sphere layers is initiated. The GaN growth step will require process conditions optimized to allow flow through the void and to prevent growth on the sphere surfaces, which would likely result in the formation of undesirable polycrystalline GaN. Figure 2 illustrates the simulated growth process through the void network formed by four close-packed sphere layers. Vertically-propagating defects are in theory blocked at each sphere layer. Some of the key technical challenges anticipated for this approach include: optimizing the process conditions such that transport of gas reactants is facile through the sphere layers, particularly for smaller-diameter spheres where the void spaces are narrower; preventing nucleation of GaN on the silica sphere surfaces, which may be difficult especially in the tightly-confined spaces between adjacent spheres; and controlling the self-assembly of the sphere layers over large areas (e.g. the number of layers).

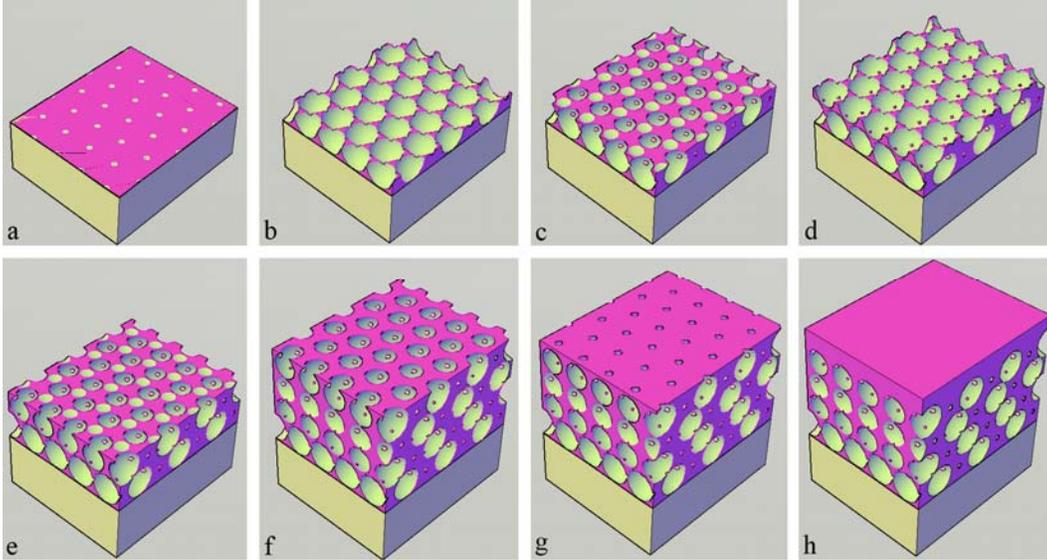


Figure 2. A series of conceptual 3D diagrams illustrate the sequence of the GaN epitaxy inside the voids between SiO₂ spheres: (a), initial GaN nucleation on a sapphire substrate with the coverage close to unity; (b), the GaN film thickness reaches the maximum radius of the SiO₂, where maximum defect blocking occurs; (c)-(e), the same process repeats at the second layer of voids; (f)-(h) the final coalescence of GaN over the top SiO₂-sphere layer.

3. Experimental Details

A ~2- μm -thick GaN layer was first deposited onto a 2-inch *c*-plane (0001) sapphire substrate by metal-organic chemical vapor deposition (MOCVD) in an Emcore D125 reactor. This “GaN-1” layer serves as a nucleation layer (similar to ELO techniques) that facilitates growth of the subsequent GaN growth through the sphere layer. This initial GaN epilayer deposition typically employed a low-temperature GaN buffer growth step at 550 °C followed by a high-temperature GaN growth at 1050 °C. For both steps, the typical flow rates of trimethylgallium and NH₃ were set to 0.107 mol/min and 10.5 standard liter/min, respectively. This growth process results in a typical TDD of $3 \times 10^9 \text{ cm}^{-2}$ in our samples, although we note that state-of-the-art growth techniques with TDDs in the mid- 10^8 cm^{-2} range for GaN films on *c*-plane sapphire using low-temperature nucleation layers have been reported.[4, 5] The samples were subsequently treated in a H₂SO₄:H₂O₂ mixture (4:1 by volume) to obtain a hydrophilic GaN surface for subsequent silica microsphere assembly.

Drip-evaporation and spin-coated methods for depositing silica sphere layers onto the substrates were initially tested and used. These methods are fast and simple, and can deposit multiple layers of spheres at once, but control over the uniformity (e.g. number of sphere layers) over wide areas is difficult. For follow-on experiments in which a monolayer of silica microspheres uniformly deposited over large areas (~ 1 inch square, the largest size possible with our Langmuir-Blodgett equipment) was desired, a Langmuir-Blodgett technique was used following a process reported by Reculosa and Ravaine[6]. Figure 3 shows uniform deposition and close packing of silica spheres over a relatively large area using this method. GaN was then selectively deposited through the sphere monolayer(s) at a typical growth temperature of $\sim 1050^\circ\text{C}$.

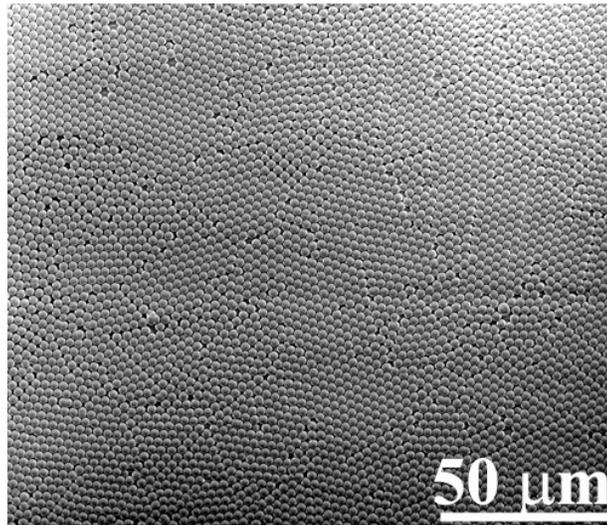


Figure 3. Close-packed monolayer of silica spheres deposited by a Langmuir-Blodgett technique, showing approximately 83% coverage (where ideal coverage = 90.7%).

Various growth conditions were tested for selective GaN regrowth through the spheres. For the optimized regrowth through a monolayer of $3\ \mu\text{m}$ diameter spheres, a “two-step” process was employed. The first step was performed at $1050\ ^\circ\text{C}$ and 450 Torr for 45 min to promote GaN facet formation. The growth pressure was then reduced to 50 Torr in the second step to facilitate lateral growth and obtain a fully coalesced film. A $2\text{-}\mu\text{m}$ -thick GaN-on-sapphire sample without silica microspheres was simultaneously loaded for the two-step regrowth as a control. Following the GaN regrowth process, the samples were treated *in-situ*

in SiH₄ at 860 °C for 2 min to decorate the threading dislocations for accurate counting by atomic force microscopy (AFM).[7] The samples were also characterized by scanning electron microscopy (SEM), x-ray diffraction (XRD), transmission electron microscopy (TEM), and cathodoluminescence (CL).

4. Results

4.1. Selective GaN growth through multiple sphere layers

By experimenting with different process conditions and sphere sizes, we were able to successfully grow GaN through silica films consisting of one or more monolayers of spheres (up to sixteen layers were successfully tested). As anticipated, penetration of GaN into layers of smaller sphere diameters is more difficult, and during initial experiments GaN growth using sphere sizes of 170 nm and two or more layers thick was not observed. By increasing the sphere diameter to 500 nm, however, we were able to grow through four sphere layers and achieve a coalesced smooth GaN film above the sphere layers. A near cross-section SEM demonstrating this successful growth is shown in Figure 4.

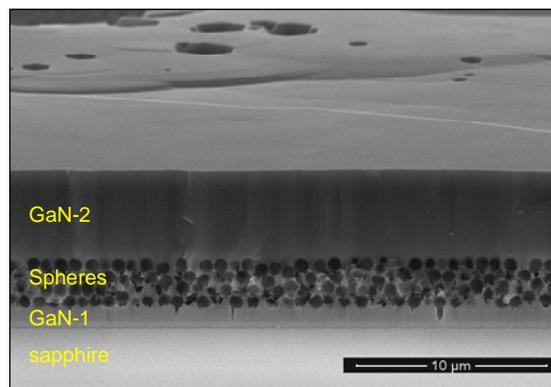


Figure 4. Side-view SEM demonstrating successful GaN growth through four sphere layers to form fully coalesced GaN.

Transmission electron microscopy (TEM) was performed in order to investigate the quality of the post-sphere “GaN-2” layer and the degree of improvement, if any, over the

underlying “GaN-1” layer. Electron-transparent slices were prepared by focused ion beam to produce cross-section samples of the GaN-1, sphere, and GaN-2 layers, and plan-view samples of the GaN-2 layer, where the GaN-2 layers were grown through roughly four layers of 500 nm diameter silica spheres. Figure 5 shows cross-section TEM images showing the sapphire substrate, GaN-1 layer, four layers of spheres, and GaN-2 layer. The growth in the GaN-2 layer is clearly single crystalline, indicating selective growth of the GaN. The TEM images show a high number of threading dislocations in the GaN-1 layer, which appear typically as dark and vertical lines, followed by a dramatic decrease in number in the GaN-2 layer after growth through the sphere layers. Quantitative analysis of the GaN-2 layer via TEM of a plan-view sample reveals that the GaN is single crystalline with an approximate dislocation density of $3 \times 10^8 \text{ cm}^{-2}$. This compares favorably with the dislocation density of the underlying GaN-1 layer, which is estimated to be $6 \times 10^9 \text{ cm}^{-2}$. This initial result represents an approximate twenty times reduction in dislocation density following growth through the four sphere layers, and thus successfully demonstrated the “proof of concept” that single crystalline GaN can selectively be grown through self-assembled 3D templates of silica spheres, with the sphere layers functioning as a defect reduction template.

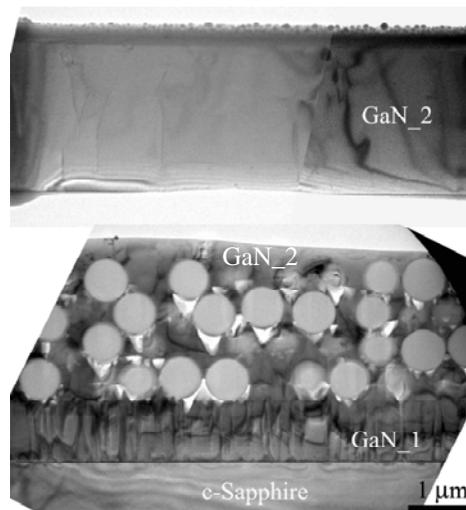


Figure 5. Cross-section TEM images showing numerous threading dislocations in the GaN on sapphire layer, followed by a drastic reduction in number in the “GaN-2” layer after growth through four layers of 500 nm silica spheres. The top cross-section was taken near the surface of the film.

4.2. Effect of sphere diameter on efficacy of technique and optimized growth

Interestingly, in general an increase in the number of sphere layers *did not improve the TDD and in most cases a single monolayer of spheres produced films with the lowest TDD*. Thus, we focused on examining the impact of sphere size on the efficacy of the technique. The degree of reduction in TDD using this technique was found to be strongly dependent on the sphere diameter, as shown in Figure 3. *Specifically, a large increase in the efficacy of the technique is observed as the sphere diameter is increased, with an impressively low TDD of $4 \times 10^7 \text{ cm}^{-2}$ achieved by using a monolayer of $3 \mu\text{m}$ spheres*. In contrast, as the size of the silica sphere is reduced to submicron scale, the TDD reduction become less significant. We note that as the sphere diameter increases from $1 \mu\text{m}$ to $3 \mu\text{m}$, the TDD scales inversely with the square of the sphere diameter, which suggests that the TDD dependence on sphere diameter is related to dislocation regeneration upon GaN coalescence over the silica spheres. We assume that the density of coalescence events, which lead to TD formation, is proportional to the density of GaN islands formed on the silica sphere template, which scales as the inverse square of the sphere diameter. Therefore, the TDD should scale roughly as the inverse square of the silica sphere diameter, which is observed. We have not yet employed sphere sizes larger than $3 \mu\text{m}$ to see if further improvements in TDD would result. We note that the use of faster growth techniques, such as hydride vapor phase epitaxy ($100\text{-}200 \mu\text{m/hr}$ vs. $1\text{-}2 \mu\text{m/hr}$ for MOCVD), may enable the practical use of larger diameter silica spheres, which require longer regrowth, and potential further reductions in the TDD.

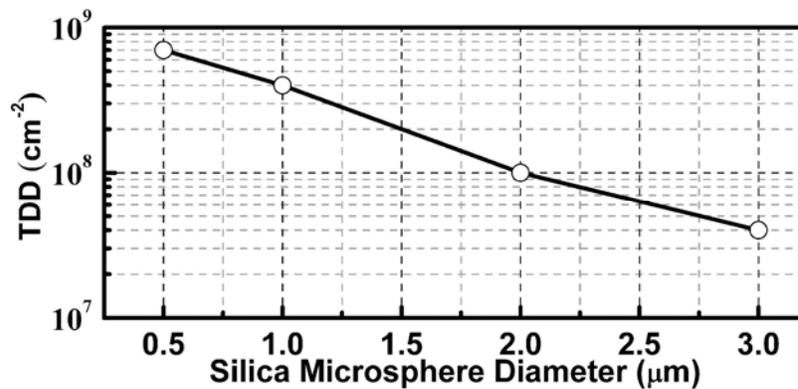


Figure 6. TDD in GaN-2 overlayers as a function of silica sphere diameter.

The GaN films grown through a single monolayer of 3 μm silica spheres was characterized in detail. The full width at half maximum of the XRD peak for GaN (0004) omega scan is 140 arcsec; this is significantly narrower than the 382 arcsec measured on the control samples regrown without the silica sphere masking layer. Figure 7 shows typical 5 \times 5 μm AFM scans of the decorated GaN surfaces. The surface of the control GaN sample is dominated by a high density of dislocations which manifest as pits, as shown in Figure 7(a). A total of 827 dislocations are counted, resulting in a TDD of $3.3\times 10^9\text{ cm}^{-2}$. In contrast, only 15 pits are counted on the GaN surface grown using the silica microsphere template, as shown in Figure 7(b), indicating a TDD in the mid 10^7 cm^{-2} range. In order to more accurately determine the TDD for the GaN film grown through the silica microsphere template, large area (27 $\mu\text{m}\times 27\ \mu\text{m}$) CL imaging was also performed, which indicated a TDD of $4\times 10^7\text{ cm}^{-2}$.

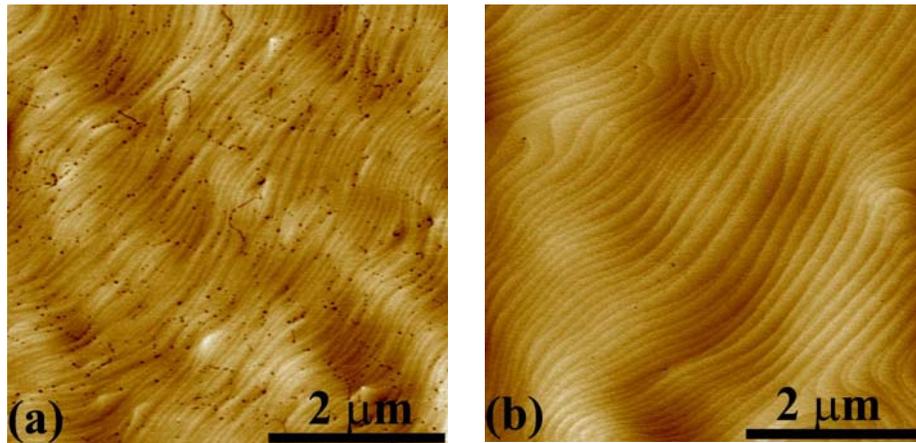


Figure 7. Typical 5 \times 5 μm AFM scans showing the surface morphology of GaN regrown without (a) and with (b) the use of a monolayer of 3 μm diameter silica microspheres.

We attribute the large TDD reduction resulting from this technique to two primary mechanisms. The first mechanism is direct dislocation blocking by the silica spheres, which is revealed in a cross-sectional TEM image in Figure 4. A large number of vertically-propagating TDs can be seen to be directly terminated by the silica/GaN interfaces. However, this direct blocking mechanism only accounts for a maximum of ~ 6 times reduction in the TDD because only $\sim 83\%$ ($5/6$) of the close-packed surface area is directly

covered by the spheres, as measured by plan-view SEM. Instead, a nearly two orders of magnitude dislocation reduction is observed in the regrowth layer. Herein, we propose that dislocation bending serves as an additional mechanism for TD reduction. In Figure 4, the TDs are shown to change their propagating directions to an angle that is nearly perpendicular to the GaN/silica interface. This phenomenon is explained by the dislocation following a path of minimum elastic energy per unit of growth length of materials.[8] The silica spheres provide a large growth interface to facilitate TD bending. This bending terminates dislocations that are not directly blocked by the spheres and may otherwise potentially propagate through the openings between neighboring spheres. In addition to these bent threading dislocations, a number of extended defects can be seen above the spheres as indicated by the arrows in Figure 4. We speculate that these defects may be dislocations that have undergone bending above and near the sphere interface, such that they are no longer vertically propagating toward the top GaN surface.

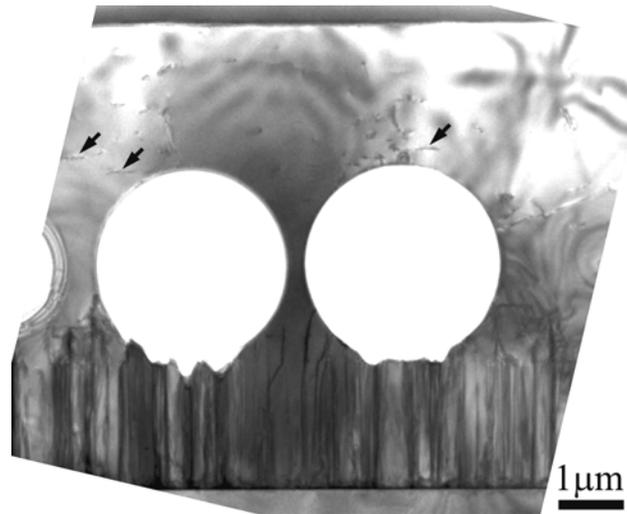


Figure 8. Cross-section TEM image showing dislocations (dark vertical lines) directly blocked and bending towards 3- μm diameter SiO_2 spheres and terminating at the GaN/ SiO_2 interface leaving a low TDD area above the spheres

4.3. Serial application of technique

A serial application of the technique, similar to a double-ELO process, was also tested in order to determine if lower TDDs could be achieved by a second regrowth step through an additional layer of SiO₂ spheres. Figure 9 shows a cross-section SEM image taken after GaN growth through an initial layer of 3 μm SiO₂ spheres, followed by Langmuir-Blodgett deposition of an additional layer of 3 μm SiO₂ spheres and subsequent GaN regrowth through this additional layer. CL images taken after the first and second regrowth steps through the sphere layers show the TDD remains unchanged at $4 \times 10^7 \text{ cm}^{-2}$, indicating serial applications of the technique do not improve the TDD. This result is also consistent with the size-dependent data and suggests that the origin of the TDDs following growth through a sphere layer are newly formed or regenerated since the majority of the TDDs from the underlying GaN layers are blocked by the sphere layer.

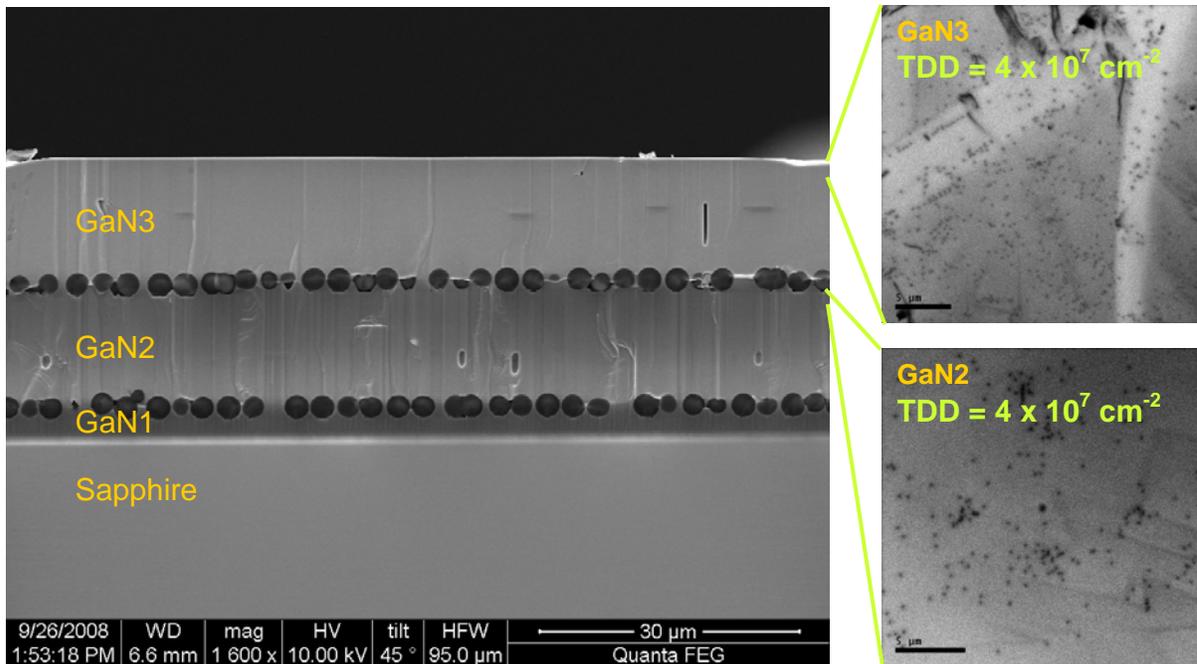


Figure 9. Cross-section SEM image of serial application of technique through two separate layers of 3 μm diameter SiO₂ spheres, with plan-view CL images taken after each growth step.

4.4. Demonstration of technique for improved GaN on Si growth

The sphere-layer TD-filtering technique was also investigated for the growth of low TDD GaN on Si(111). Growth of GaN on Si substrates has been heavily pursued due to the inexpensive cost and large size of Si wafers and the potential for integrating GaN and Si-based devices. However, the large lattice mismatch (~17%) and particularly large thermal expansion coefficient mismatch (~56%) between GaN and Si causes high tensile stress in the GaN which leads to even higher TDDs than for GaN on sapphire growth as well as crack formation upon cooling.

For the GaN on Si(111) growths, a ~30 nm thick AlN layer was first grown on the Si(111) surface via MOCVD, followed by a ~0.5 μm thick gradient AlGaIn layer with GaN at the top of the layer. A single layer of 3 μm silica sphere was coated on the surface using the Langmuir-Blodgett technique as described before. A GaN regrowth was performed using the same optimized growth conditions used on GaN covered sapphire substrates, the results of which are shown in Figure 10. The TDD on the sphere covered regions was measured by large-area CL scans to be $\sim 6 \times 10^7 \text{ cm}^{-2}$, an extremely large improvement in comparison with the control sample (no silica spheres applied), where the TDD was estimated to be in the $\sim 10^{10} \text{ cm}^{-2}$ range. Cracking was also noticeably reduced in the GaN layer grown through the sphere layer in comparison to the control sample, indicating a reduction in thermal-strain effects.

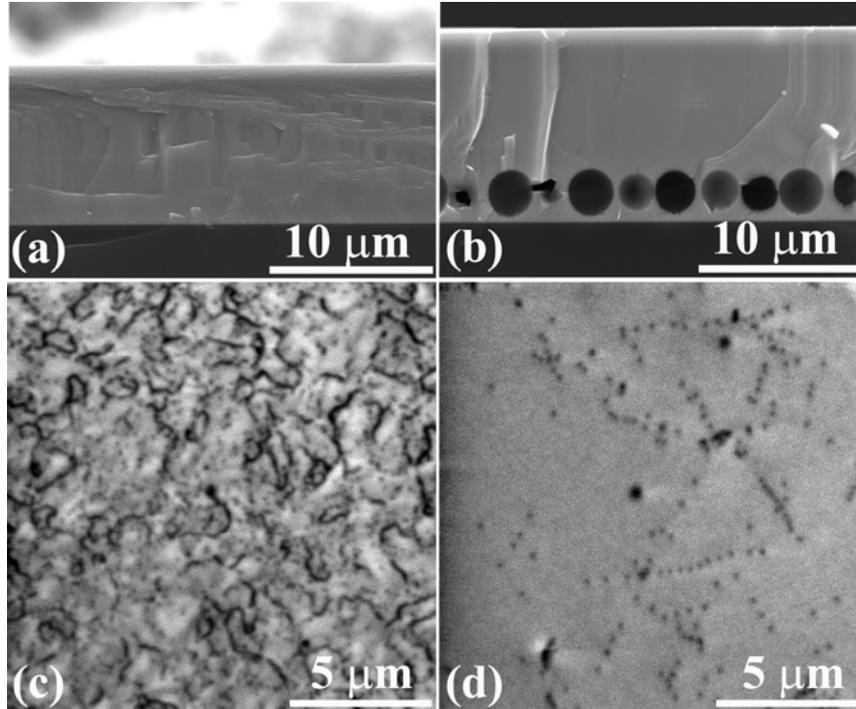


Figure 10. SEM cross-section image of GaN on Si(111) growth (a) without spheres, and (b) through a layer of 3 μm spheres. Plan-view CL images of the GaN surface (c) without spheres, and (d) through spheres, showing large reduction in TDD.

5. Conclusions & Future Work

A simple technique for growing high quality GaN on 2-inch sapphire with very low TDDs of $4.0 \times 10^7 \text{ cm}^{-2}$ by using self-assembled silica microspheres as a dislocation filtering dielectric growth mask was demonstrated. Compared to conventional epitaxial lateral overgrowth schemes, this technique does not require expensive lithographic patterning steps or equipment. The sphere diameter was found to have a significant impact on the dislocation reduction, with larger spheres resulting in lower dislocation densities, presumably due to a lower density of coalescence interfaces. Dislocation blocking and bending are attributed to the significant reduction of dislocations.

In the future, we anticipate that this method may be readily extended to other GaN orientations (e.g. nonpolar GaN) and substrates, as well as other mismatched heteroepitaxial materials systems (particularly growth on Si substrates), since the dislocation blocking and bending mechanisms are not inherent to one particular system. Further improvements in the

technique may be realized by using larger sphere sizes and by further optimization of the growth conditions.

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Appendix – List of Publications & Patent Applications

Refereed Publications:

Q. Li, J. J. Figiel, G. T. Wang, "Dislocation density reduction in GaN by dislocation filtering through a self-assembled monolayer of silica microspheres", *Applied Physics Letters* **94**, 231105 (2009).

Technical Advances/Patent Applications:

Q. Li and G. T. Wang, "LOW-DISLOCATION-DENSITY EPITATIAL LAYERS GROWN BY DEFECT FILTERING BY SELF-ASSEMBLED LAYERS OF SPHERES" SD10594, U.S. Patent Application 12/388,103 filed on February 18, 2009.

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