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GaN quantum dot density control by rf-plasma molecular beam epitaxy

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We report on the growth of GaN quantum dots and the control of their density in the Stranski–Krastanov mode on AlN (0001) by rf-plasma molecular beam epitaxy at 750 °C. After depositing the equivalent of 2–3 ML GaN coverage, as limited by N fluence under Ga-droplet growth conditions, excess Ga was desorbed and Stranski–Krastanov islands formed under vacuum. We present the dependence of island density as a function of GaN coverage (for two growth rates: 0.10 and 0.23 ML/s), as estimated from atomic force microscopy and cross-sectional transmission electron microscopy. With a GaN growth rate of 0.23 ML/s, the island density was found to vary from less than 3.0×10^8 – 9.2×10^{10} cm⁻² as the GaN coverage was varied from 2.2 (critical thickness) to 3.0 ML. For a GaN growth rate of 0.10 ML/s, the island density varied from 2.0×10^{10} to 7.0×10^{10} cm⁻² over a GaN coverage range of 2.0–3.0 ML. For each growth rate, the GaN islands were found to be of nearly uniform size, independent of the quantum dot density. © 2004 American Institute of Physics. [DOI: 10.1063/1.1645333]

The control of GaN quantum dot (QD) size and density is of great interest for the fabrication of QD devices and investigation of their physical properties. Low-density, size-controlled GaN QDs are necessary for the study of spatially isolated QDs by techniques such as cathodoluminescence and microphotoluminescence without the need for metal masks or etched mesas. Higher densities of GaN QDs are potentially useful to improve the efficiency of active regions for optoelectronics applications for emission of ultraviolet to blue light.

Several groups have demonstrated control of GaN QD growth by metalorganic chemical vapor deposition (MOCVD) and molecular beam epitaxy (MBE).^{1–4} In rf-plasma MBE of GaN on AlN (0001) at 750 °C, it has been shown that above 2.5 ML coverage, GaN will transition from a coherently strained two-dimensional (2D) layer to the Stranski–Krastanov (SK) configuration [coherently strained 2D wetting layer plus three-dimensional (3D) islands].³ Simultaneous size and limited density control has also been demonstrated; small, high-density SK islands have been ripened under N flux or vacuum to produce larger, low-density islands.⁴ Recently, significant progress has been made toward understanding the surfactant effect of excess Ga in thin film GaN growth (under a dynamically stable Ga adlayer) on AlN (0001) by rf-plasma MBE.^{3,5} Despite recent progress in control of the growth of GaN QDs by rf-plasma MBE, the lowest reported density of MBE-grown GaN QDs is 4

$\times 10^{10}$ cm⁻² at 3 ML of GaN coverage.³ In this work, we report growth of GaN QDs by plasma-assisted MBE with densities from 3.0×10^8 to 9.2×10^{10} cm⁻² via control of N-limited GaN coverage prior to the SK transition. We show that the density of GaN QDs may be varied, nearly independent of mean GaN QD size, via N-fluence control under Ga-rich conditions.

Growths were performed in an Applied EPI Gen II MBE system, equipped with conventional Al and Ga Knudsen effusion cells and a standard Unibulb nitrogen rf-plasma sources. We used *ex situ* high-resolution x-ray diffraction of GaN/Al_{0.1}Ga_{0.9}N superlattices grown on ~ 2 - μ m-thick MOCVD-GaN template to calibrate the N-limited GaN growth rate corresponding to 150 W rf-plasma forward power and 0.2–0.4 sccm nitrogen flow. Radial (ω - 2θ), triple-axis x-ray diffraction scans about GaN (0002) were compared to dynamical simulations⁶ to correlate layer thickness, grown under Ga-droplet conditions, to deposition time. The N-limited growth rate uncertainty ($\pm 1\%$, typical) determined for each superlattice sample by this method was less than the overall fluctuation of the growth rate ($\pm 1.9\%$) for separate sample growths within the two growth rates used in this study.

GaN QDs were grown at 750 °C by rf-plasma MBE on partially relaxed 100 nm AlN buffers on ~ 2 - μ m-thick MOCVD-GaN on sapphire. AlN was grown under slightly metal-rich conditions ($f_{\text{Al}}/f_{\text{N}} \sim 1.1$). Before GaN deposition, excess Al was consumed under N flux exposure (i.e., producing AlN). The AlN surfaces were simultaneously exposed to Ga and N flux ($4 < f_{\text{Ga}}/f_{\text{N}} < 8$), in the Ga-droplet regime at 750 °C.⁷ Under these conditions, the nominal GaN coverage

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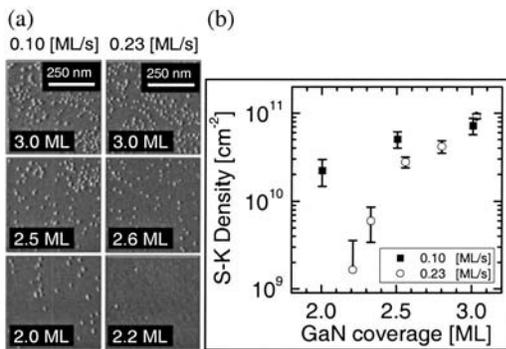


FIG. 1. SK island density for various GaN coverages for two GaN growth rates. (a) AFM (amplitude) micrographs of GaN SK surfaces. (b) GaN SK island density as a function of GaN coverage. Error bars indicate standard deviation from separate measurements.

was specified by the supplied active N flux, which is understood to diffuse rapidly on or through the excess Ga ad layer(s). The morphological evolution of the surfaces was then observed via reflection high-energy electron diffraction (RHEED) along the $[11\bar{2}0]$ azimuth under vacuum. Above 2.0 ML nominal GaN coverage, we observed the concurrent desorption of excess Ga and the onset of the GaN SK transition after 5–20 s as evidenced by the RHEED transition from 2D diffraction surface normal streaks to 3D diffraction Bragg spots.^{3,8} The samples were either cooled immediately and studied by atomic force microscopy (AFM), or overgrown with AlN for examination by cross-section transmission electron microscopy (TEM).

We used AFM to characterize GaN SK islands on partially relaxed AlN buffers. In a series of ~ 1 cm² samples for two growth rates (0.10($\pm 1.9\%$) and 0.23($\pm 1.9\%$) ML/s), we observed a strong correlation between island density and GaN coverage from 2.0 to 3.0 ML (Fig. 1). Independent of growth rate, the island density saturated at $\sim 9 \times 10^{10}$ cm⁻² at 3.0 ML GaN coverage. Below 3.0 ML coverage, the 0.10 ML/s growth rate series had higher island densities than the 0.23 ML/s series. The opportunity for increased N diffusion (under excess Ga) prior to the SK transformation at the lower growth rate may provide for enhanced diffusion of adatoms to stable island nucleation sites. This effect may account for higher island density at a given coverage for the lower growth rate series. The difference between the island density trends for the two series may also be a result of Ga fluence. The Ga flux (1×10^{-6} beam equivalent pressure) and substrate temperature (750 °C) were held constant for both growth rate series; the 0.10 ML/s series was exposed to approximately two times greater Ga fluence during the GaN deposition under Ga-droplet conditions. Under our growth conditions, the GaN surface was initially smooth under adsorbed excess Ga, and subsequently evolved into the SK configuration as the excess Ga desorbed. The delay after GaN deposition, but before the SK transition, during which N is able to diffuse rapidly, may be critical in the transition from the (metastable) coherently strained 2D layer to the partially relaxed SK configuration.

Within the limits of AFM measurement, the mean GaN SK island heights and diameters were nearly independent of the island density for each growth rate. As the GaN coverage was varied from 2.2 to 3.0 ML (GaN growth rate: 0.23 ML/s

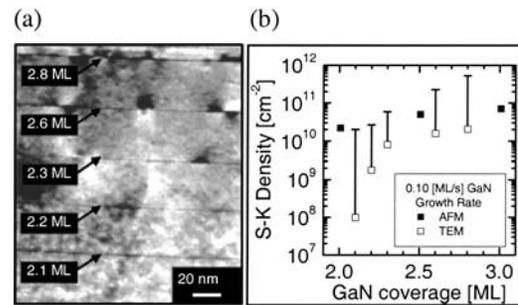


FIG. 2. (a) Cross section TEM micrograph of a stack of GaN SK islands (QDs) with varying GaN coverage in AlN (0001). (b) Estimated density of SK islands as a function of GaN coverage by AFM and TEM. Error bars are one standard deviation.

s), the mean island height exhibited a modest increase (12%), from 2.5 ± 0.7 to 2.8 ± 0.6 nm. Over the same range of GaN coverage, the SK island height distributions remained large (standard deviations 20%–30% of mean height) independent of island density. The mean island diameter (full width at half maximum) for the 0.23 ML/s series was also nearly independent of coverage, exhibiting an overall mean of 11 ± 7 nm. For the lower growth rate (0.10 ML/s) series, the mean island height was 2.4 ± 0.7 nm, and the mean island diameter was 16 ± 7 nm. The QD sizes and densities obtained from AFM measurements were used to estimate the amount of GaN contained in the SK islands, assuming a lens (spherical cap) island shape. For both growth rates, the amount of GaN contained in the islands was equivalent to the excess (above critical thickness for the SK transition) GaN deposited. This indicates that after GaN deposition under Ga droplet conditions, the excess Ga desorbs, but the supplied N fluence remains on the surface during the SK transition. The critical thickness for the 0.23 ML/s series was 2.2 ML, and for the 0.10 ML/s series the critical thickness was apparently less than 2.0 ML. Our results indicate a GaN (2D) “wetting layer” of approximately 2 ML is formed initially, followed by the formation of SK islands with excess GaN.

To verify the control of GaN QD density (GaN SK islands immediately covered by AlN) as a function of GaN coverage, a multilayer stack of GaN QDs was examined by two-beam TEM. The GaN coverage was varied from 2.1 to 2.8 ML, (growth rate: 0.10 ML/s), with 30 nm AlN spacer layers. The dependence of QD density on coverage was consistent with the relationship found on the uncapped GaN SK surfaces (Fig. 2).

For moderate to high GaN coverage (above 2.5 ML), the surfaces of individual ~ 1 cm² samples exhibited nearly uniform island density. The surface of a ~ 1 cm² sample (growth rate 0.23 ML/s; coverage 2.2 ML) had a maximum island density of 1.2×10^{10} cm⁻² and a minimum density of 3×10^8 cm⁻². A contour plot of this surface (Fig. 3) was generated by the use of a correlation method to grid nine individual island density data points into a 3×3 matrix. The island density *decreased* in the direction defined by the impinging N plasma flux. The amount of available N (sticking coefficient ~ 1 under excess Ga conditions)^{7,9} for GaN growth is expected to decrease along the direction of N impingement, consistent with our correlation of SK island density to GaN coverage as determined by the supplied active

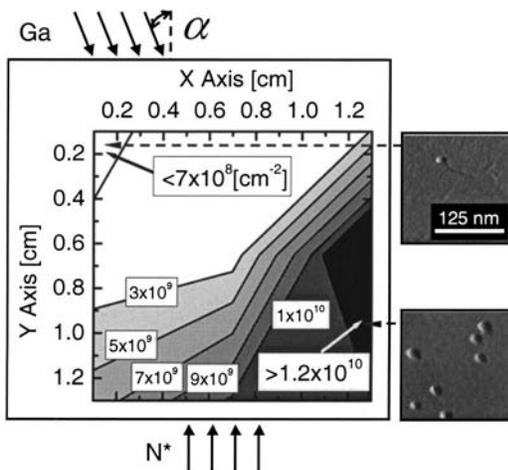


FIG. 3. SK island density on the surface of a 2.2 ML GaN coverage sample (GaN growth rate: 0.23 ML/s). Approximate Ga and N* flux sample-plane impingement directions are shown schematically ($\alpha=45^\circ$). Representative AFM (amplitude images) depict the highest and lowest island density regions.

nitrogen. The SK island density *increased* in the direction defined by the impinging Ga flux, consistent with the proposed role of Ga as a surfactant for GaN growth on AlN (0001).⁵ On this sample, the lowest island density ($3 \times 10^8 \text{ cm}^{-2}$) was observed near the highest Ga flux and the lowest N flux, while the highest island density ($1.2 \times 10^{10} \text{ cm}^{-2}$) was observed near the lowest Ga flux and the highest N flux.

In conclusion, we have demonstrated a route for control of GaN QD density over more than two orders of magnitude, by control of the GaN coverage and growth rate. We found that the GaN QD density may be controlled nearly independent of QD size.

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