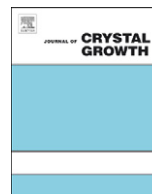




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Selective area growth of *a*- and *c*-plane GaN nanocolumns by molecular beam epitaxy using colloidal nanolithography

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ABSTRACT

Selective area growth of *a*-plane GaN nanocolumns by molecular beam epitaxy was performed for the first time on *a*-plane GaN templates. Ti masks with 150 nm diameter nanoholes were fabricated by colloidal lithography, an easy, fast and cheap process capable to handle large areas. Even though colloidal lithography does not provide a perfect geometrical arrangement like e-beam lithography, it produces a very homogeneous mask in terms of nanohole diameter and density, and is used here for the first time for the selective area growth of GaN. Selective area growth of *a*-plane GaN nanocolumns is compared, in terms of anisotropic lateral and vertical growth rates, with GaN nanocolumns grown selectively on the *c*-plane.

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The main advantages of one-dimensional structures such as nanocolumns, compared to thin films, is the dislocation- and strain-free growth on different substrates. This higher crystal quality yields a better efficiency in devices such as light emitting diodes (LEDs) based on InGaN/GaN quantum disks (QDisks) [1–4].

Much effort has been dedicated to the growth of self-assembled nanocolumns (NCs) by Plasma-Assisted Molecular Beam Epitaxy (PAMBE), allowing a better knowledge of the material physical properties and growth mechanisms [5–8]. However, the strong morphology dispersion, typical of a self-assembled process, hinders both the processing of nanodevices arrays and their electrical behavior (NCs merging, defect generation, current injection inhomogeneities). Indeed, arrays of self-assembled nanoLEDs show in general much lower electroluminescence efficiency than the corresponding one measured by photoluminescence (PL). In addition to that InGaN active regions embedded in self-assembled NCs of different diameters and lengths always show fluctuations in the composition, leading to multicolor emission. Arrays of nanostructures grown by selective area growth (SAG) provide a much better homogeneity in terms of morphology, electrical and optical characteristics. SAG of

III-nitride NCs [9–12] and other nanostructures [13,14], is generally performed on very thin metal (Ti, Mo) or dielectric (SiO₂, SiN) masks with an array of nanoholes.

A relevant issue concerning optoelectronic devices based on III-nitrides is the presence of strong polarization fields that may reduce efficiency. This is the case in layers grown along the *c*-axis and, a huge effort is nowadays dedicated to the growth of high quality non-polar and semi-polar material [15], with a particular emphasis on non-polar LEDs [16].

In the case of SAG of GaN NCs on *c*-plane GaN templates, the growth front is generally formed by semi-polar (*r*-planes) that yield a “pencil-like” profile [12]. This profile is then transferred to InGaN QDisks embedded within the GaN NC. Though in this structure the effects of internal fields in the active region of the device (i.e. nanoLED) may be reduced compared with polar planes, the most effective solution is to grow along non-polar directions, either by growing core-shell heterostructures on the lateral nonpolar facets of otherwise polar NCs, or by growing axial heterostructures along nonpolar directions such as [10–10] (*m*-planes) or [11–20] (*a*-planes). In both approaches, the internal fields are nominally zero.

This work reports on the SAG of *a*-plane GaN NCs (i.e. the growth direction is parallel to [11–20]) by PAMBE on Ti masks. The vertical and lateral growth rates (anisotropy) are studied and compared with *c*-plane GaN NCs SAG.

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Nanohole masks are fabricated by colloidal lithography [17–19] on *a*-plane and *c*-plane GaN templates. Colloidal solution is formed by polystyrene (PS) nanobeads (260 nm of average diameter and sulfate covered, so negatively charged, and purchased at Invitrogen). Commercial solution is then diluted in water at 4% w/v. On the other side, GaN templates were alternately dipped in poly(sodium 4-styrenesulfonate) (PSS) and poly(diallyldimethylammonium chloride) (PDDA) solutions (2% w/v, during 1 min each step), which are negatively and positively charged poly-electrolytes, respectively. Poly-electrolytes are purchased at Sigma-Aldrich. A tri-layer PSS/PDDA/PSS (~5 nm thick) is formed to achieve a negatively charged (like the nanobeads surface) and hydrophilic surface. The GaN/PSS/PDDA/PSS surface is covered with the colloidal solution by spin-coating (2000 rpm for 30 s). The result is a monolayer of PS nanobeads, with a close-packaged arrangement in large areas (domains up to $400 \times 400 \mu\text{m}^2$). Once the nanobeads are arranged on the surface, reactive ion etching (RIE) oxygen plasma (40 W; oxygen pressure of 80 mTorr, 1 nm/s) is used in order to reduce the diameter of the beads and, consequently, to isolate them. After the plasma etching, 7 nm of Ti is evaporated on the surface and a lift-off process results in an array of 150 nm diameter nanoholes on the Ti layer covering the GaN surface. The colloidal lithography process is depicted in Fig. 1.

During the PAMBE growth (Riber Compaq 21) the substrate temperature was monitored with a thermocouple located at the growth stage. The Ga and N fluxes were calibrated in equivalent (0001) GaN growth rate units for compact layers (nm/min) [7,12,20,21]. Scanning electron microscopy (SEM) images were obtained with a CRESTEC CABL-9500C system. Atomic force microscopy (AFM) images were taken with a Nanoscope microscope (MMAFM-2) and processed with Nanotec software WS \times M [22]. Based on these measurements the growth rates were estimated. Also transmission electron microscopy (TEM) measurements in high resolution (HREM) and selected area electron diffraction modes (SAED) were carried out in a JEOL2010F and a Jeol2100 instruments respectively, in order to confirm the crystal orientation.

Previously to the GaN SAG, a nitridation step was performed following the process reported in Ref. [10,12] to prevent Ti mask

degradation at high temperature. Growth time was either 1 h or 3 h at 880 °C with $\Phi_{\text{Ga}}=16 \text{ nm/min}$ and $\Phi_{\text{N}}=5 \text{ nm/min}$ or 10 nm/min. Nominal flux ratios ($\Phi_{\text{C}}/\Phi_{\text{N}}$) were always larger than one, but due to the high growth temperatures used for SAG the actual $\Phi_{\text{C}}/\Phi_{\text{N}}$ ratio at nanoholes (growth front) was significantly reduced [12].

Fig. 2 shows SEM images of SAG GaN NCs along the *c*-plane (Fig. 2(a and b)) and *a*-plane (Fig. 2(c and d)), grown under the same conditions: 1 h at 880 °C, $\Phi_{\text{Ga}}=16 \text{ nm/min}$ and $\Phi_{\text{N}}=5 \text{ nm/min}$. In both cases, perfect selectivity was achieved, but morphology differences were clearly observed. *c*-Plane GaN NCs had a typical hexagonal structure with pyramidal top surfaces (pencil-like), as expected from previous results [9–14]. However, nanostructures grown on *a*-plane substrates did show elongated geometries with several lateral facets, as observed by SEM and AFM (inset in Fig. 2(c)). The vertical growth rates measured for *c*- and *a*-plane SAG were of 3.5 nm/min and 1.3 nm/min, respectively. In fact, the measured ratio between both growth rates (2.7) is in the range of the factor derived from the kinetic Wulff's plots (ν -plots) calculated for GaN in Ref. [23] (factor 4).

In order to check the morphology evolution of the *a*-plane nanostructures with time and growth conditions, two new samples were grown: (a) like that on Fig. 2(c and d) (880 °C, $\Phi_{\text{Ga}}=16 \text{ nm/min}$ and $\Phi_{\text{N}}=5 \text{ nm/min}$) but during 3 h, and (b) during 1 h but under higher Φ_{N} (10 nm/min). Both samples did show similar morphologies (Fig. 3(a and b)) evolving from elongated (Fig. 2(c)) to arrow-headed shape, similar to those reported by MOVPE [23]. The main difference is that the sample with higher Φ_{N} showed the onset of the nanostructures coalescence. Fig. 3(c–e) shows magnified images with the evolution sequence. It is worth to mention the strong lateral growth anisotropy observed in the *a*-plane nanostructures. Regarding the height of the nanostructures in both cases, either increasing the active nitrogen by a factor of two, or the growth time by a factor of three, led to a proportional increase of height.

A thin foil TEM lamella has been prepared using focused ion beam (FIB). The cross-section was obtained across the direction perpendicular to that of the elongation of the nanocolumns (it is orthogonal to the arrow in Fig. 3(e)). Fig. 4(a) presents a general

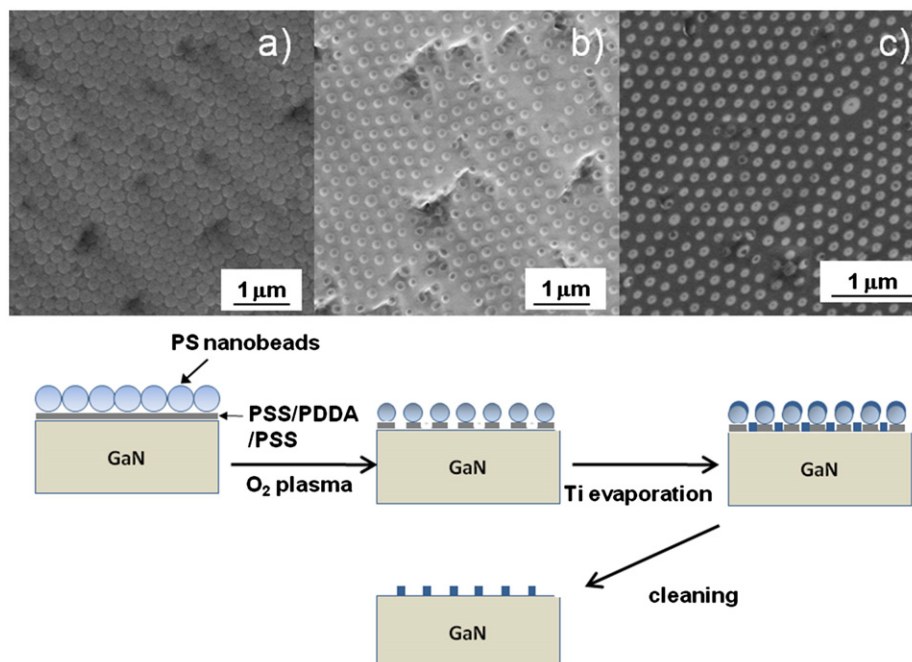


Fig. 1. Colloidal lithography scheme for a 7 nm thick Ti mask with 150 nm diameter nanoholes on a GaN template. SEM pictures show: (a) close-packaged arrangement of PS nanobeads; (b) after oxygen plasma etching and (c) the final Ti mask.

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