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Electrical and structural properties of GaN films and GaN/InGaN light-emitting diodes grown on porous GaN templates fabricated by combined electrochemical and photoelectrochemical etching

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ABSTRACT

Porous GaN templates were prepared by combined electrochemical etching (ECE) and back-side photo-electrochemical etching (PECE), followed by the overgrowth of GaN films and InGaN/GaN multiple quantum well (MQW) light-emitting diode (LED) structures. Structural, luminescent, and electrical properties of the GaN and LED structures were studied and compared with the properties of structures grown under the same conditions on templates not subjected to ECE–PECE treatment. Overgrowth of LED structures on the ECE–PECE templates reduced strain, cracking, and micropits, leading to increased internal quantum efficiency and light extraction efficiency. This luminescence enhancement was observed in overgrown GaN films, but was more pronounced for InGaN/GaN LED structures due to suppression of piezoelectric polarization field in QWs.

1. Introduction

GaN and related ternary and quaternary compounds are currently widely used for fabrication of UV–visible light-emitting diodes (LEDs) and laser diodes (LDs), of high electron mobility transistors (HEMTs), power p–i–n diodes and Schottky diodes, photodetectors operating in a wide spectral range, efficient solar batteries, and many other devices (see e.g. Refs. [1–10]). In many such devices there are certain advantages in growing the structure by heteroepitaxy on one type of substrate and then separating it and transferring to another substrate. For example, all devices could benefit considerably if grown on cheap and readily available Si substrates allowing integration with Si-based controlling electronics and capitalizing on good thermal conductivity of Si (see e.g. [4,11]). High-power electronic devices require very good thermal conductivity and would benefit tremendously if grown on diamond substrates [6]. However, the crystalline quality of GaN structures grown on Si and diamond is inferior to the quality of the structures grown on sapphire. Hence, developing methods of separation of GaN-based films and heterostructures from sapphire substrate and transferring the free-standing films to other substrates of choice is a very important task.

In GaN technology several methods based on wet etching have been developed to solve this task. They are all based on having this or that kind of sacrificial layer underlying the structure of interest and etching this sacrificial layer selectively to facilitate lateral separation. The main problems are attaining the high lateral etching rate to allow separation of large-area films and preserving the high device quality of the separated portion. The former is limited by efficient delivery of the etching agent to the etch front, the latter by one's ability to not introduce a high density of defects in the active layer of the device. Unfortunately, no uniformly applicable methods of solving these tasks have been developed so far. For example, electrochemical etching (ECE) has been successfully employed in Refs. [12–14] for chemical lift-off (CLO) of LED GaN-based structures. In this approach a small chip of GaN is predefined by dry etching, with the low-temperature buffer layer underlying the chip exposed to lateral ECE. The obvious disadvantage of the method is the low lateral etch rate coming from very limited access of the etchant to the interface. The result is that only small chips can be separated and transferred that way. (A somewhat different approach was successfully tried in Ref. [15] where a thin InGaN film underlying the active GaN layer to be separated was introduced. Backside illumination through the sapphire substrate in

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combination with photoelectrochemical etching made possible lateral undercutting of the GaN by etching only the InGaN underlayer. Selectivity was achieved by employing a GaN filter during electrochemical etching under illumination because of the lower bandgap of GaN. Small chips of about 100 mm in diameter produced by dry etching could be separated that way. Obviously, separation of large diameter pieces or chips with GaN/InGaN quantum wells should present a problem in such technique.) The improvement of the ECE method proposed in Refs. [16–18] overcomes this constraint, but preserving the high quality of the separated layers and their suitability for subsequent overgrowth requires very careful optimization. In Ref. [18] the impact of the porous GaN template prepared by ECE on mechanical properties of overgrown GaN was studied as a function of oxidation conditions. Effects of the underlying porous GaN template on electrical properties and electroluminescence efficiency of GaN/InGaN LED structures were reported in Ref. [19]. In our earlier work [20] we proposed a method combining ECE approach with subsequent photoelectrochemical etching (PECE) and GaN film overgrowth to provide a template for fabrication of easily detachable free-standing GaN layer. However, there remained an uncertainty as to the method being capable of preserving or improving the quality of thus fabricated layers or device structures compared to the standard growth techniques. In what follows we demonstrate that such GaN films and multi-quantum-well (MQW) GaN/InGaN LED structures indeed have the structural, electrical, and optical properties either of the same quality or superior in several aspects to the properties of the structures similarly grown by metalorganic chemical vapor deposition on sapphire.

2. Experimental

The combined process of ECE–PECE can be briefly described as follows. One started with growing by MOCVD on basal plane sapphire of a two-layer template consisting of approximately 1 μm thick unintentionally doped uid-GaN film followed by approximately 2 μm thick n^+ (concentration of around 5×10^{18} cm^{-3}) GaN layer. This two-layer composition was then first subjected to the EC etching in oxalic acid (0.2 M) at room temperature [20]. The *n*-type GaN and a platinum wire were used as the anode and cathode, respectively. The EC etching was performed at 15 V for 30 min. As a result a porous GaN film with the pores diameter of about 40 nm was formed. The pores were approximately aligned along the [0001] growth direction and extended down to the uid-GaN that served in this ECE process as an etch-stop, because the etch-rate drops down dramatically with the decrease in donor concentration in GaN (see e.g. [21–23]) (in our case this residual donor concentration was below 10^{16} cm^{-3}, see e.g. Ref. [24]). At the second stage back-side photoelectrochemical etching in potassium hydroxide solution in water (KOH, 0.06 M) was carried out at 9 V for 30 min. The 300 W ultraviolet OSRAM lamp (non-filtered) was used as the light source and the Keithley voltage/current source (model No. 2400) was used to set the voltage and control the current during the process. Because dislocations in GaN are very effective lifetime-killers [25,26] the concentration of photogenerated holes that act as the main oxidation agents in this PECE process was greatly diminished in the vicinity of dislocations in the uid-GaN subjected to etching. Consequently, PECE proceeded rapidly mostly in the dislocation-free parts of the uid-GaN, thus leaving behind the columnar structure with the nanorods density on the order of dislocation density (around 10^9 cm^{-2} in our case). The great advantage of the combined ECE–PECE approach is that the channels formed in the n^+-GaN layer during the ECE stage serve for efficient delivery of the etchant to the reaction front at the PECE stage and thus provide the rapid and uniform formation of the nanocolumn structure across the entire wafer. This ECE–PECE processed GaN template then was used for regrowth of either n$^+$ (5×10^{18} cm^{-3} Si donors) GaN single films with thickness of 4 μm at 1060 °C or of the entire multi-quantum-well (MQW) LED structure consisting of 4-μm-thick n^+GaN contact layer ($n = 5 \times 10^{18}$ cm^{-3}), 5 GaN/InGaN quantum wells (QWs), and 0.15-μm-thick p-GaN top contact layer. Undoped GaN barrier and InGaN well were grown at temperatures of 850 °C and 750 °C, respectively. For comparison similar single layer n^+-GaN and MQW LED reference structures were grown on the two-layer uid-GaN (1 μm)/n^+-GaN (2 μm) templates not subjected to the ECE–PECE processing.

The ECE–PECE template and the ECE–PECE overgrown and reference samples were characterized by scanning electron microscope (SEM) imaging, by room temperature microcathodoluminecence (MCL) spectra measurements, by MCL imaging using either panchromatic MCL signal or monochromatic MCL with the chosen wavelength. Structural characterization involved triple-axis high-resolution X-ray (HRXRD) measurements and MCL imaging. SEM experiments were performed using field emission scanning electron microscopy (FE-SEM, JEOL, S-4700). Panchromatic MCL imaging was done using LEO 1450 VP SEM with the probing beam current of 200 pA. MCL spectra measurements and MCL spectral imaging were performed with higher beam current of (1–3) nA. Experimental setups for SEM characterization are described in detail in our earlier paper in Ref. [27]. HRXRD measurements were performed for the (0002) reflection using the triple-axis Bede D1 system in the ω–θ rocking curves and the θ–2θ modes [28].

In addition, the MQW LED structures were processed into actual LEDs with 300 μm × 300 μm pads by photolithography and dry etching. For the reference and ECE–PECE template LEDs room temperature capacitance–voltage *C–V*, room temperature current–voltage *I–V*, and room temperature light output versus driving current *L–I* measurements were performed as described in Ref. [29].

3. Results and discussion

3.1. ECE–PECE template structure and properties

As mentioned above, the starting ECE stage in treating the double layer template gave rise to the formation of pores approximately aligned along the [0001] film growth direction. The pores went down to the interface with the uid-GaN film and had the average pores diameter close to 40 nm and the porosity of about 18%. Subsequent PECE processing led to the formation of additional pores, to slight increase of the average pores diameter to 42 nm with the increased porosity of 27%, to breaking the alignment of some of the pores along the [0001] direction and to the formation of nanopillar structure around the dislocations in the uid-GaN portion of the template (see Fig. 1(a) showing the SEM plan-view image after ECE, Fig. 1(b) for the plan-view image after ECE–PECE, and Fig. 1(c) for the cross-sectional SEM image of the template after the ECE–PECE processing). The columnar structure formed in the uid-GaN film is expected to serve as a weak spot along which

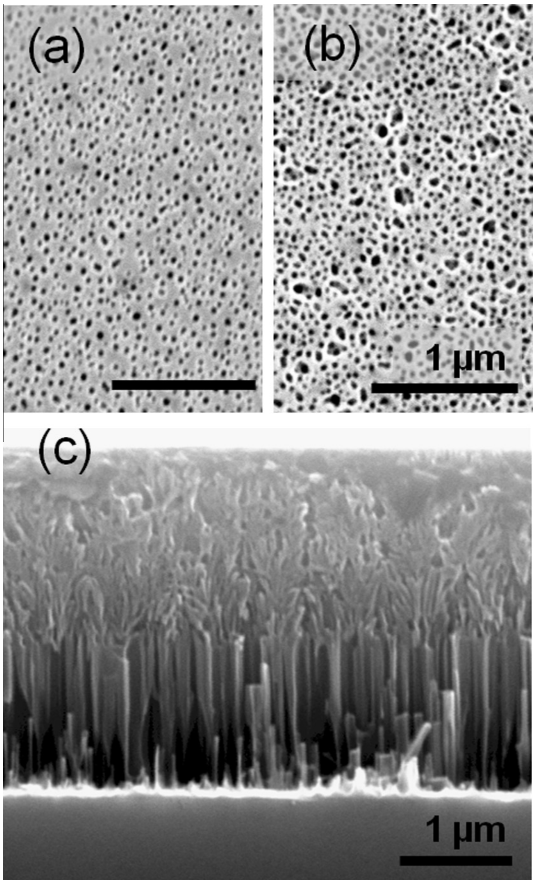

Fig. 1. SEM image of the template after ECE (a), the template after ECE–PECE (b), and cross sectional SEM image of the ECE–PECE template (c).