Bragg polariton luminescence from a GaN membrane embedded in all dielectric microcavity

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We report on the development of a band gap-selective photochemical etching technique capable of producing 200 nm thick optical quality freestanding GaN membranes. The use of low electrolyte concentration combined with intense excitation by a laser source are shown to yield highly anisotropic etch profile with optical quality etched surfaces. Using this technique, high quality GaN microcavity is fabricated by embedding the GaN membrane inside an all-dielectric mirror cavity. In these structures, Bragg polariton photoluminescence is observed at room temperature. © *2011 American Institute of Physics*. [doi:10.1063/1.3595481]

Wide band gap based semiconductor microcavities, operating in the strong exciton–photon coupling regime have been shown to be very promising candidates for low threshold lasing at room temperature.[1,2] The achievement of strong coupling regime in GaN based system requires high cavity finesse and relatively narrow excitonic resonances. However, monolithic growth of GaN based microcavities is extremely challenging due to the large difference in lattice constants between III-nitride binaries, resulting in build up of mismatch strain and cracking of the epitaxial structures.[3] Employing lattice matched to GaN III-nitride alloys, for distributed Bragg reflector (DBR) fabrication, in order to address this problem, restricts the refractive index contrast and consequently the width of the mirror stopband, which becomes comparable to the Rabi splitting.[4,5] To bypass these limitations, recent reports have focused on the fabrication of fully dielectric microcavity structures, by either selective wet etching of sacrificial layers[6–8] or mechanical polishing of thicker GaN films to produce membranes.[9,10] Such structures have been used to demonstrate polariton anticrossing at room temperature, however fabrication challenges remain considerable, particularly in relation to the optical smoothness requirements of the GaN membrane surfaces.

In this work, we extend the previously reported resonantly enhanced photoelectrochemical (PEC) etching technique[11] in *lateral direction*, to achieve highly selective etching of a sacrificial InGaN layer with an anisotropy ratio better than 1:2000. We show that this technique can be used to fabricate high optical quality freestanding 200 nm thick GaN membranes exhibiting narrow excitonic photoluminescence (PL) at room temperature. Integration of these high quality membranes into all-dielectric microcavity structures allows the observation of Bragg polariton luminescence at room temperature.

The sample used in selective PEC etching experiments is shown in Fig. 1(a). It consists of a 200 nm GaN film on top of a 50 nm $In_{0.1}Ga_{0.9}N$ sacrificial layer, epitaxially grown, by plasma assisted molecular beam epitaxy (PA-MBE) on commercial 3 μm thick unintentionally doped (0001) GaN on sapphire template. The sacrificial layer had a thickness below the critical one and it was fully coherent with the GaN substrate, as confirmed by x-ray diffraction. The sample was patterned using standard lithographic process into a grid of 40 μm$\times$40 μm mesas, shown in Fig. 1(b). The mesas, with height equal to 1 μm, were fabricated by reactive ion etching (RIE), in order to expose laterally the sacrificial $In_{0.1}Ga_{0.9}N$ layer for subsequent selective PEC etching.

The PEC etching was carried out in an electrochemical cell at room temperature, using several concentrations of KOH solution in a setup described in detail in Ref. 11. An indium Ohmic contact was used to apply a reverse dc bias of 4 V to the semiconductor, required for the confinement of photogenerated holes at the electrolyte/semiconductor interface. The selectivity of the PEC etching process was ensured by the resonant excitation of the sacrificial $In_{0.1}Ga_{0.9}N$ layer at a wavelength below GaN band gap, using a 405 nm laser diode source, while the concentration of the electrolyte solution was varied by two orders of magnitude.

In Fig. 2, scanning electron microscopy (SEM) images after the PEC etch are shown, for different KOH concentrations of 0.01*M*, 0.1*M*, 1*M* employed, under constant bias. Etch time was 1 h for the first two concentrations and 15 min for the last one. It should be noted that the power density of

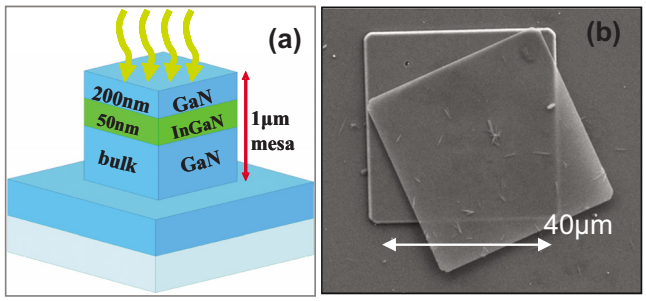

FIG. 1. (Color online) (a) schematic sample structure and (b) SEM picture of detached GaN membrane lying on the underneath mesa.

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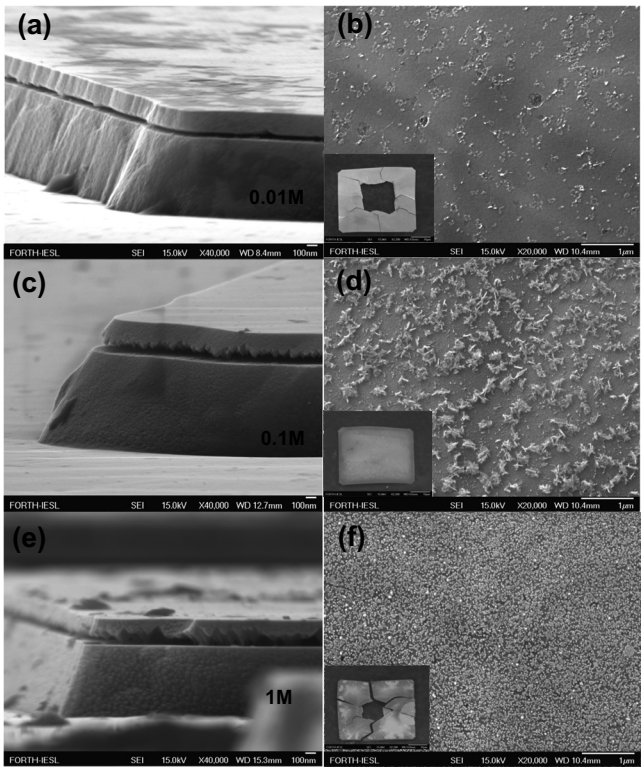

FIG. 2. [(a), (c), and (e)] SEM images of undercuts achieved after the removal of the $In_{0.1}Ga_{0.9}N$ sacrificial layer using 0.01*M*, 0.1*M*, 1*M* KOH concentrations respectively, and [(b), (d), and (f)] zoomed-in SEM images of the corresponding N-face surfaces revealed after etching. The insets show zoomed-out images of the complete membranes after transfer on carbon tape. In cases where the etch process is incomplete the membrane cracks during the transfer process revealing the edges of the etch front, (b) and (f), from which the etch rates can be readily extracted as shown in Fig. 3.

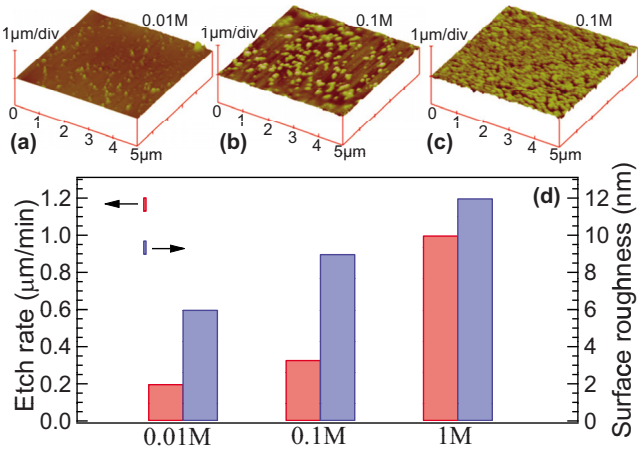

FIG. 3. (Color online) AFM images of N-face membrane surface following lateral PEC etching with KOH concentrations of 0.01*M* (a), 0.1*M* (b) and 1*M* (c). (d) Histogram of etch rate (left column) and rms surface roughness (right column) for the specified concentrations, showing clear correlation between etch rate and surface quality.

0.32 W/cm^2, delivered to the sacrificial layer by band gap selective excitation, is two orders of magnitude higher than in previous reports.[12–15] The highly selective nature of the etching process, with anisotropy of at least 1:2000 (estimated lower limit obtained from close-up SEM analysis of the membranes), ensures the clear undercut of the sacrificial $In_{0.1}Ga_{0.9}N$ layer while the top GaN membrane is subject to negligible thinning. Following etching, the membranes are transferred onto a carbon tape and the quality of the underlying N-face GaN surface is carefully examined by SEM and atomic force microscopy (AFM). As seen in Figs. 2(b), 2(d), and 2(f) and Figs. 3(a)–3(c), the bottom N-face GaN membrane surface exhibits etch byproducts which decrease in density with decreasing KOH concentration. At lowest KOH concentration, the surface is nearly free of byproducts (independently confirmed by energy dispersive x-ray spectroscopy), as shown in Figs. 2(b) and 3(a), producing optically smooth etched surfaces with ~6 nm rms roughness.

On the contrary, increased roughness of the etched surface and cone-shaped morphology is obtained with increasing KOH concentration, in agreement with previous reports, which used broadband excitation and similar electrolyte concentrations. Notably, for high KOH concentrations the N-face of GaN appears chemically unstable yielding a cone shaped surface. The dependence of etch rates and surface roughness on KOH concentration, extracted from SEM and AFM images, is presented in the histograms of Fig. 3(d). A clear correlation between the surface quality and KOH concentration is found, showing that a fine interplay between etching rate and selectivity is required for the optimization of the etched surface quality, which is improved at low KOH concentrations.

The above results are significant, since they suggest that, by carefully tuning the etch conditions, it is possible to achieve band gap dependent etching selectivities better than 1:2000, in the InGaN/GaN material system. The relatively low KOH electrolyte concentrations in combination with the high optical power and spectrally narrow excitation source in this study, enhance the band gap selective nature of the lateral PEC etching process.

Once the etching conditions are optimized we proceed to the fabrication of a complete all-dielectric microcavity structure. We start by fabricating mesa structures of different sizes, which are formed by RIE using ten period SiO_2/Ta_2O_5 dielectric DBR as a hard mask. Evaporation of the DBR mask protects the top side of the GaN membrane throughout the PEC process. Figure 4(a) shows a SEM image of the GaN membrane attached to dielectric DBR mirror, following lateral PEC etching. It reveals a high quality etched surface, as well as individual layers of the dielectric DBR mirror. Following the PEC etch, membranes are transferred onto a carbon tape, without any damage, provided the etching front has progressed throughout the sacrificial layer. Evaporation of the second dielectric DBR mirror completes the fabrication process of the microcavity structure as shown in Fig. 4(b).

Linear optical properties of the fabricated microcavity structures at room temperature are investigated by collecting PL spectra at different angles using an optical fiber. For this purpose, a continuous wave laser at 325 nm is focused on a 3 μm diameter spot through a long working distance microscope objective, exciting the sample nonresonantly. In Fig. 4(c), normalized PL spectra in the range of 0°–68° are presented. For small angles, clear excitonic emission is observed, coinciding with the PL emission from a bare GaN membrane shown in the graph on the right. At larger angles, PL emission arises from filtered excitonic PL through photonic Bragg modes on the low energy side of the mirror stopband. As confirmed from reflectivity measurements on reference samples and as expected theoretically, the photonic Bragg modes of both DBR mirrors, split into distinct TE

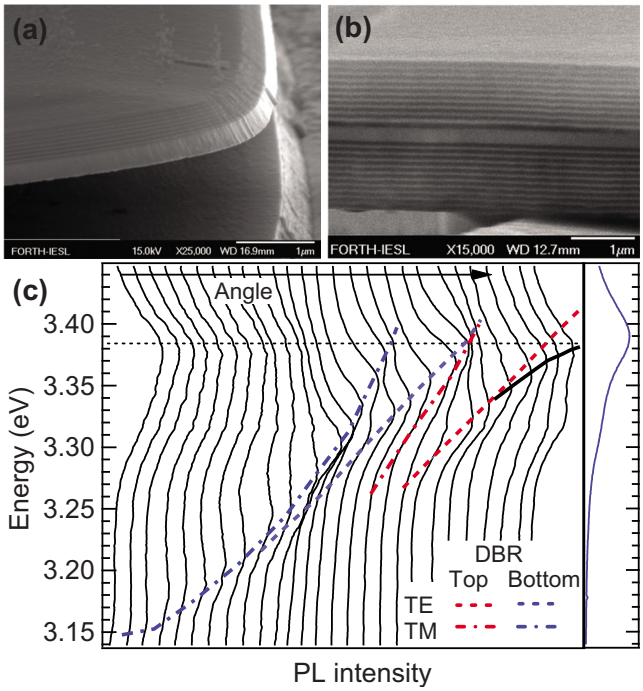

FIG. 4. (Color online) (a) SEM image of detached GaN membrane after PEC etch coated with dielectric mirror on the top side. (b) Complete all-dielectric GaN microcavity structure following carbon tape lift-off and evaporation of second dielectric DBR. (c) Intensity normalized PL spectra from GaN microcavity sample taken within angular range of 0°–68°. PL spectra from a bare GaN membrane is also shown for reference (graph on the right). TE (TM) Bragg modes of top (bottom) DBRs are marked in dashed (dotted) lines and are just guide for the eye. Bragg-polariton luminescence is observed at large angles (solid black line).

(TM) modes above 15°, as shown in Fig. 4(c) with dashed (dotted) lines. It should be noted that, the bottom DBR stopband is shifted by design to shorter wavelengths by ~10 nm to allow transmission of the laser excitation at 405 nm required for PEC etching. Good agreement between PL emission spectra and modeled DBR mirror Bragg modes is independently confirmed up to 60°.

However, at larger angles there is a clear discrepancy between the observed PL emission and the top DBR TE Bragg mode dispersion. Instead of crossing, Bragg mode, gradually takes the position of excitonic resonance, a behavior characteristic of the strong coupling regime. Such exciton-Bragg mode coupling is known to produce two new eigenmodes of the system called Bragg polaritons.[16] Notably, the anticrossing behavior is observed only for the TE Bragg mode of the top DBR mirror having the highest cavity finesse values at such large angles while being in the vicinity of the excitonic resonance. However, the upper Bragg polariton branch cannot be clearly resolved due to the presence of bulk GaN continuum states above excitonic transition. We also note that the present sample does not exhibit the conventional exciton-cavity mode strong coupling regime since for the specific membrane thickness the sample is positively detuned at normal incidence.

Bragg polaritons have been observed previously in CdTe based microcavities,[17] owing to large exciton oscillator strength in this material. However until now, unavailability of high quality GaN microcavities with wide mirror stopbands has prevented such observation in GaN microcavities. Owing to its relative technological maturity and its proven capability of room temperature operation, GaN microcavities are expected to play a leading role in the future realization of ultraefficient polariton based devices.

In conclusion, we show that by careful optimization of the PEC etching conditions, it is possible to produce high selectivity lateral etching of buried InGaN layers in a GaN matrix. Using this band gap selective lateral etching technique, ultrathin optical quality freestanding GaN membranes were fabricated and sandwiched between dielectric mirrors to form a microcavity. In these high quality microcavities, strong exciton-Bragg mode coupling is observed opening exciting opportunities for exploration of polariton nonlinearities such as lasing, parametric amplification, and oscillation at room temperature.[18]

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