Thin Solid Films 627 (2017) 77–81

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

# Fabrication and whispering gallery resonance of self-rolled up gallium nitride microcavities

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ARTICLE INFO

*Article history:*
Received 20 October 2016
Received in revised form 11 February 2017
Accepted 25 February 2017
Available online 01 March 2017

*Keywords:*
GaN
Microtube
Rolled-up
Whispering gallery mode (WGM)

ABSTRACT

The *C*-plane monocrystalline GaN/ZnO thin film epitaxially grows on *A*-plane sapphire substrate by molecular beam epitaxy (MBE) is revealed by the reflection high-energy electron diffraction (RHEED). The monocrystalline GaN nanomembranes have been rolled up into tubular microstructures as whispering gallery microcavities, which support the whisper gallery modes (WGMs) in the violet and blue regime. The WGMs of the rolled-up monocrystalline GaN devices are tunable with microtube diameter by tuning the strain gradient of the GaN nanomembranes. This approach could help with not only the further development of GaN-based photonic devices, but also the physical understanding of other rolled-up optical microcavities.



## 1. Introduction

Gallium nitride (GaN), as one direct bandgap semiconductor with excellent optical properties, acts as the leading material for the application for optoelectronic devices [1], particularly ultraviolet (UV) and visible micro-/nanoscale light sources operating at room temperature [2–6]. However, the progress of GaN-based optical microdevices is still demanding the fabrication of high quality freestanding GaN layers and enabling them to be shaped and assembled into new structures and devices [6–9]. Recently, UV-assisted electroless chemical etching [1], chemical lift-off [10], in-situ lift-off [11,12], or laser lift-off [13] have been also proposed to achieve high quality free-standing GaN layers or membranes. However, the residual strains during growth and process could lead to dislocations and macroscopic cracks [11,14,15]. Hence, effective strain-engineering can help the preparation of high-quality GaN-based nanobelts [16] or high quality free-standing GaN wafer (350 μm) [17]. Recently, strain-engineering is also adopted for shaping single/composite nanomembranes into size-scalable three-dimensional (3D) architectures due to the elastic energy minimization [18,19]. Rolled-up micro- and nanoscale tubular optical cavities can be realized by standard photolithography and etching process based on the design of sacrificial layers [18]. The luminescent spectra and the light confinement in such ring-like optical resonators are tunable by modifying the functional layer materials, designing the complex 3D geometry and surface modification [8,18]. Various interesting materials were used for the fabrication of self-rolled up tubular optical microcavities at difference spectral range. As for oxide self-rolled up optical microcavities, their luminescent spectra in yellow and red region are attributed to defect center, such as $SiO_x/Si$ [20], $SiO_x/SiO_2$ [21], $Y_2O_3/ZrO_2$ [22], and $TiO_2$ [22], etc. (Fig. 1). The high index semiconductor self-rolled up microcavities based on InGaAs quantum wells (InGaAs QW) [23–25] and PbS quantum dots (PbS QD) [26] show typical whisper gallery modes (WGMs) with high quality-factor (Q-factor) and 3D light confinement in near infrared region (Fig. 1). Recently, the WGMs with high Q-factor of luminescent $Cd_6P_7$ nanoparticles embedded $TiO_2$ microtube cavities extend from the visible to the near infrared [27]. Although previous theoretical and experimental results on GaN-based rolled-up micro-/nano-tubes have largely focused on the design of the tube structure [7,8,28], there is no report on rolled-up tubular optical microcavities based on GaN material for the resonance in the range of violet and blue.

In this work, we develop an effective process for fabricating GaN-based tubular optical microcavities by the rolling of single crystal GaN nanomembranes. The typical photoluminescence spectra of self-rolled up GaN microtube at room temperature indicated that these GaN microtubes support the WGMs in the violet and blue regime. We fabricate tubular GaN optical microcavities with WGMs through the self-rolled up nanotechnology by releasing of thin *c*-oriented GaN epilayer from *A*-plane sapphire substrate with a sacrificial ZnO layer. Our approach hints an interesting method to build up GaN-based photonic devices.

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http://dx.doi.org/10.1016/j.tsf.2017.02.059

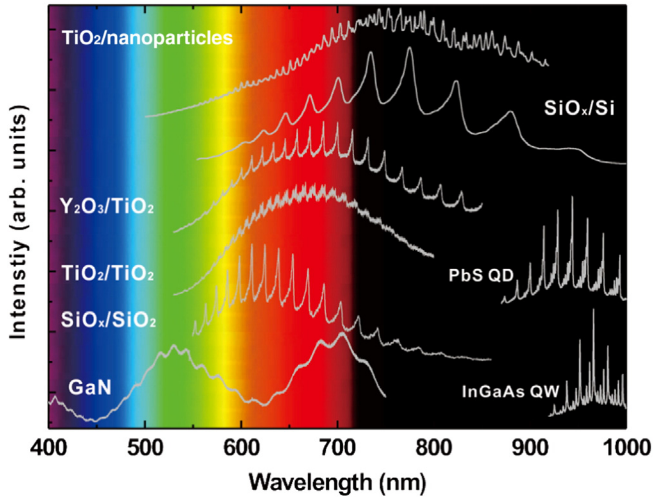


**Fig. 1.** The micro-photoluminescence (μ-PL) spectra of rolled-up microtubes in various color regions, which are composed of $SiO_x$/Si (Q < 100) [20], $SiO_x$/$SiO_2$ (Q ~ 100) [21], $Y_2O_3$/$ZrO_2$ (Q > 1500) [22], $TiO_2$ (Q > 1500) [22], InGaAs QW (Q > 2000) [23,24], PbS QD (Q > 1000) [26], $TiO_2$/nanoparticles (Q > 1200) [27] and GaN (this work, Q ~ 100).

## 2. Experimental procedure

### 2.1. Synthetic procedures

Self-rolled-up tubular GaN optical microcavities with WGMs are fabricated by releasing of thin GaN epilayer from *A*-plane sapphire substrate with a ZnO sacrificial layer. ZnO layer (i.e. sacrificial layer) and GaN film were grown by plasma-assisted molecular beam expitaxy (MBE). A ZnO buffer layer is grown directly on *A*-plane sapphire substrates at 380 °C. The oxygen ($O_2$) flow rate and plasma power are kept constant at 4 sccm and 380 W, respectively. Then, the high quality ZnO epilayer (~180 nm) are grown at 700 °C with growth rate of 2.8 nm $min^{-1}$. The oxygen ($O_2$) flow rate and plasma power are kept constant at 6 sccm and 400 W, respectively. A GaN layer is then grown on top of the high quality ZnO layer at 780 °C with nitrogen ($N_2$) flow rate at 3 sccm and plasma power at 350 W. In order to construct self-rolled-up GaN optical microcavities, the ZnO layer were employed as a sacrificing layer. In fact, the ZnO layer can be easily etched by HCl (1.7 mol $L^{-1}$) or KOH (4 mol $L^{-1}$) solutions.

### 2.2. Sample characterization

The crystal structural quality of the samples is studied in situ by reflection high-energy electron diffraction (RHEED), and by high resolution X-ray diffraction (HRXRD) and transmission electron microscope (TEM) techniques after the growth. The morphologies of GaN microtubes are detected by scanning electron microscopy (SEM) and optical microscope. The optical properties of the fabricated microtubes were tested by the micro-photoluminescence ($\mu$-PL) setup at room temperature.

## 3. Results and discussion

The sample composed of the layer sequence of sapphire substrate, ZnO and GaN from bottom to top. The fabrication process of tubular self-rolled up micro-cavities is schematically displayed in Fig. 2a. We design our experiments as following: the *c*-oriented ZnO layer can be selectively removed, releasing the active GaN layer, since the intrinsic stress gradient existing in the GaN active nanomembrane can cause it to self-assemble into a micro-tubular cavity as shown in Fig. 2a. Our sample started with the ideal *A*-plane of sapphire substrate, which is twofold symmetric, while the *C*-plane of ZnO and GaN is six-fold symmetric. However, in such systems, single crystalline thin films can be epitaxially grown by the so-called domain matching epitaxy (DME), where integral multiples of major lattice planes match across the film-substrate interface [15,29]. For GaN and ZnO layers, the 4-fold of the GaN or ZnO a lattice constant fits perfectly to the *c* lattice constant of sapphire, three GaN or ZnO $(1\bar{1}00)$ planes fit to two sapphire $(1\bar{1}00)$ planes in other direction [15,30,31]. Thus, *c*-oriented ZnO layer and GaN layer can be grown on the *A*-plane sapphire substrate.

The surface reconstructions of ZnO and GaN observed by reflection high-energy electron diffraction (RHEED) are adopted to monitor the ZnO buffer layer and GaN thin film surface during growth. Fig. 2b and c show RHEED patterns observed at the surface of the GaN/ZnO film

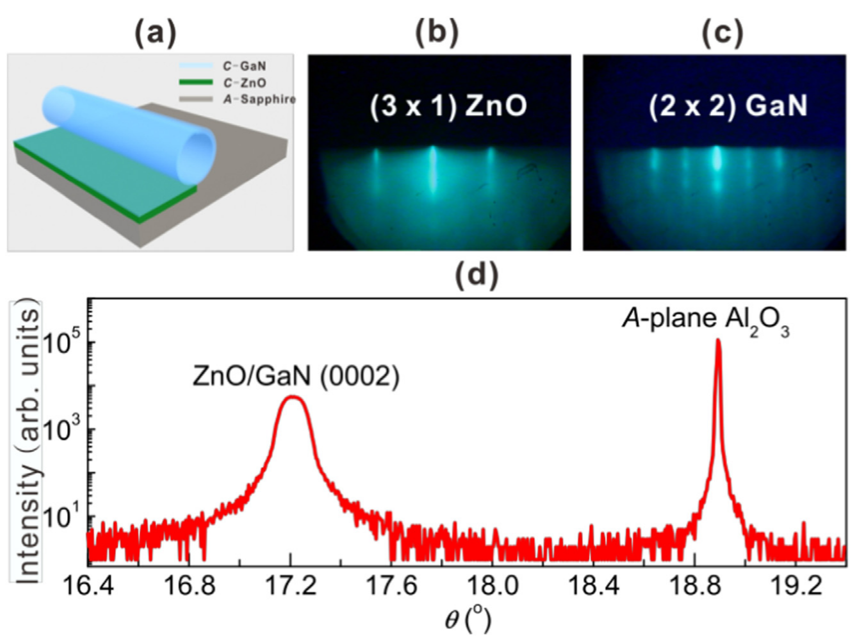


**Fig. 2.** (a) Schematic diagram illustrating the fabrication process of a rolled-up *C*-plane GaN tubular microcavity on *A*-plane sapphire substrate. RHEED pattern show the surface reconstruction of (b) ZnO and (c) GaN. (d) high resolution X-ray diffraction (HRXRD) spectra of the as-grown GaN film on ZnO/*A*-plane sapphire substrate. All the diffraction angle are calibrated by the *A*-plane sapphire substrate $(11\bar{2}0)$.