Thin Solid Films 519 (2011) 2508–2512

Contents lists available at ScienceDirect

Thin Solid Films

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

High quality *a*-plane GaN films grown on cone-shaped patterned *r*-plane sapphire substrates

Y.Q. Sun [a], Z.H. Wu [a,*], J. Yin [a], Y.-Y. Fang [a], H. Wang [a], C.H. Yu [a], Xiong Hui [a], C.Q. Chen [a], Q.Y. Wei [b], T. Li [b], K.W. Sun [b], F.A. Ponce [b]

[a] Wuhan National Laboratory for Optoelectronics, College of Optoelectronic Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074, China
[b] Department of Physics, Arizona State University, Tempe, AZ 85287-1504, USA

ARTICLE INFO

Article history:
Received 11 May 2010
Received in revised form 19 November 2010
Accepted 1 December 2010
Available online 8 December 2010

Keywords:
Nonpolar gallium nitride
Patterned sapphire substrate
Metal-organic chemical vapor deposition
X-ray diffraction
Transmission electron microscopy
Cathodoluminescence

ABSTRACT

Planar nonpolar $(11\overline{2}0)$ *a*-plane GaN films have been grown by metalorganic chemical-vapor deposition directly on cone-shaped patterned *r*-plane sapphire substrates (PRSS) fabricated by dry etching. High-resolution X-ray diffractometers 2θ-ω scan confirmed that the films grown on PRSS are solely *a*-plane oriented, and the full width at half maximum values (FWHM) of the X-ray rocking curves for $(11\overline{2}0)$ GaN along $[0001]_{GaN}$ and $[1\overline{1}00]_{GaN}$ were found to be 684 and 828″, respectively. As compared to the film grown on conventional *r*-plane sapphire substrate which typically has $(11\overline{2}0)$ omega FWHM values of 900 and 2124″ along $[0001]_{GaN}$ and $[1\overline{1}00]_{GaN}$ respectively, the film grown on PRSS exhibits overall reduced omega FWHM values, and much smaller anisotropy behavior of crystallinity with respect to the in-plane orientation. The surface morphology is also improved by utilizing the PRSS technique. Cross-sectional transmission electron microscopy analysis shows that the density of threading dislocations has been greatly reduced from $\sim 1.0\times10^{10}$ cm^{-2} above the flat sapphire regions to $\sim 1.0\times10^{7}$ cm^{-2} above the protruding cone patterns. The improvement of crystal quality and the increase of light extraction efficiency by using cone-shaped PRSS technique lead to a strong enhancement in the light emission of *a*-plane GaN films. These results indicate that growth of *a*-plane GaN films on cone-shaped PRSS shows promise for use in high-quality and high-cost-performance nonpolar GaN based devices.

1. Introduction

Wurtzite GaN films grown along the [0001] direction have been widely used for InGaN-based light-emitting diodes (LEDs) operating in the visible range. However, optical emission from the quantum wells in such devices exhibits the quantum-confined Stark effect, causing large red-shift of the emission peak and a reduction in transition probability [1–3]. Epitaxial growth of GaN along nonpolar directions (directions normal to the [0001] direction) is expected to circumvent this problem by removing the internal fields normal to the InGaN quantum wells, which are inevitably present in the standard [0001] growth direction [4–6]. As the bulk nonpolar GaN is not readily available, heteroepitaxy of nonpolar GaN films on large-mismatch substrates such as sapphire and SiC is usually employed. However, such nonpolar films typically exhibit poor surface morphology and crystalline quality, due to the large growth anisotropy along different in-plane directions, and the formation of stacking faults and threading dislocations (TDs) that originate at the substrate interface [5,7,8]. Therefore, the defect density in the epitaxial layer needs to be reduced to improve the performance of nonpolar nitride-based LEDs. Various techniques have been proposed to reduce the defect density for nonpolar GaN, among which the epitaxial lateral overgrowth (ELO) technique with mask pattern forming steps is most commonly used [9–12]. Although such ELO technique using mask pattern can effectively improve the crystallinity of the overgrown layer, it significantly increases the manufacturing complexity due to additional growth interruption for deposition of dielectric mask. On the other hand, growth of GaN on patterned sapphire substrate (PSS) is maskless and growth interruption-free and can improve the device performance by simultaneously reducing the TD density and enhancing the light extraction efficiency [13–17]. Therefore, the PSS technique has already been extensively applied for the growth of high performance *c*-plane GaN based LEDs. However, this technique has not received adequate attention for development of nonpolar GaN based LEDs.

In this work, we have grown planar *a*-plane GaN on cone-shaped patterned *r*-plane sapphire substrates (PRSS) with hexagonal symmetry using single-step growth without mask by metalorganic chemical-vapor deposition (MOCVD). The structural, morphological and optical characteristics of nonpolar *a*-plane GaN films grown on cone-shaped PRSS have been investigated and compared to those grown on conventional *r*-plane sapphire substrate (CRSS). The results

* Corresponding author. Tel.: +86 27 87793024; fax: +86 27 87793024.
E-mail address: zhihao.wu@mail.hust.edu.cn (Z.H. Wu).

0040-6090/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2010.12.005

show that the film grown on PRSS has much improved crystallinity and superior optical properties as compared to that grown on CRSS.

2. Experimental details

Fabrication of the cone-shaped PRSS in this work was accomplished by following processes. A SiO_2 layer was first deposited on a 2-inch *r*-plane sapphire substrate by the plasma-enhanced chemical vapor deposition as the etching mask and was patterned by photolithography. Then, the cone shape of photoresist was formed using a thermal reflow technique. After the cone-like profile was transferred to SiO_2 by a reactive ion etcher, the *r*-plane sapphire substrate was then etched by utilizing an inductively coupled plasma etcher with BCl_3 plasma. Fig. 1 shows the scanning electron microscope (SEM, FEI Sirion 200) image of the cone-shaped PRSS, taken at an operating voltage of 10 kV. The pattern is uniform in size and arranged in hexagonal symmetry. The diameter and the closest spacing of the cone are both 3.0 μm. The height of each cone is 1.5 μm.

The *a*-plane GaN films were grown on the cone-shaped PRSS as described above and also on conventional *r*-plane sapphire substrate (CRSS) for comparison in a water-cooled vertical MOCVD reactor at low pressure (5.3×10^3 Pa) by using triethylgallium (TEG), and NH_3 gases as the source materials for Ga and N, respectively. First, the substrate was thermally cleaned and nitridized under NH_3/H_2 mixture at a temperature of 960 °C. Then, a thin GaN buffer layer was deposited at a temperature of 600 °C using TEG and NH_3 gases. Growth of main GaN layer on CRSS was performed at 1020 °C, and the sample thickness was around 3.0 μm. In the case of the growth on PRSS, the main layer was grown at a higher temperature of 1050 °C to facilitate film coalescence, and a fully coalesced sample was about 6.0 μm. The orientation and crystal structure of *a*-plane GaN samples were investigated by X-ray diffraction (XRD, Philips Panalytical X'pert PRO MRD) with the Cu Kα1 line ($\lambda = 1.54056$ Å) and Ge (004) used as the source and monochromator, respectively. The surface morphology of the samples was examined by a differential interference contrast microscopy (Olympus-BX51) and an atomic force microscopy (AFM, Veeco NanoScope MultiMode) in a contact mode. Cathodoluminescence (CL) was performed to evaluate the optical properties of the sample in the SEM (JEOL JSM 6300) equipped with a Gatan monoCL2 spectrometer. Cross-sectional transmission electron microscopy (TEM) samples were prepared using standard mechanical polishing and argon-ion milling techniques, and were analyzed using a field-emission TEM (Philips CM200) operated at 200 keV.

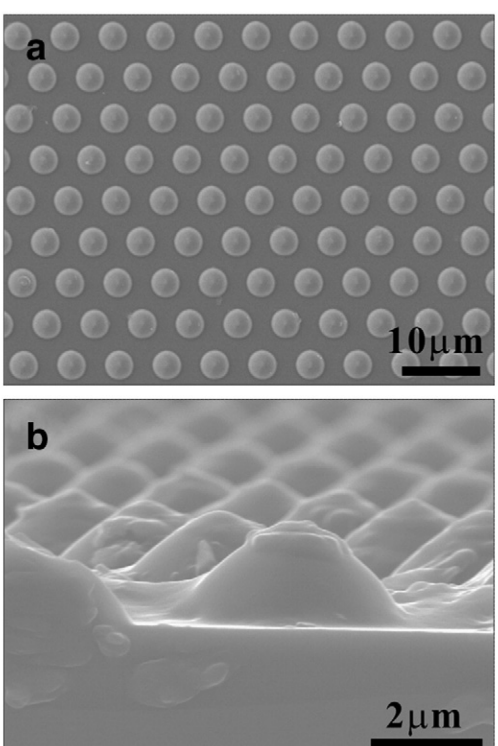

Fig. 1. SEM image of the cone-shaped patterned sapphire substrate. The diameter and the closest spacing between each cone are both 3.0 μm. The height of each cone is 1.5 μm.

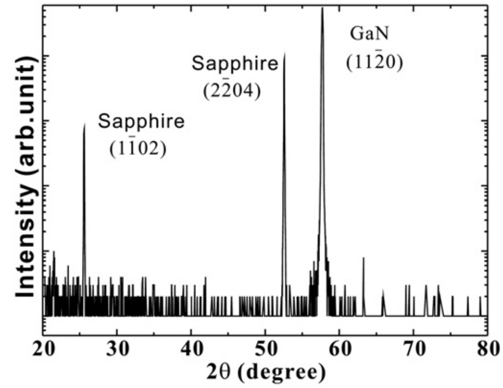

Fig. 2. Typical XRD 2θ-ω scan profile of *a*-plane GaN grown on a cone-shaped patterned sapphire substrate.

3. Results and discussion

Fig. 2 shows the typical XRD 2θ-ω profile of the *a*-plane GaN film grown on cone-shaped PRSS, taken with the X-ray incident plane

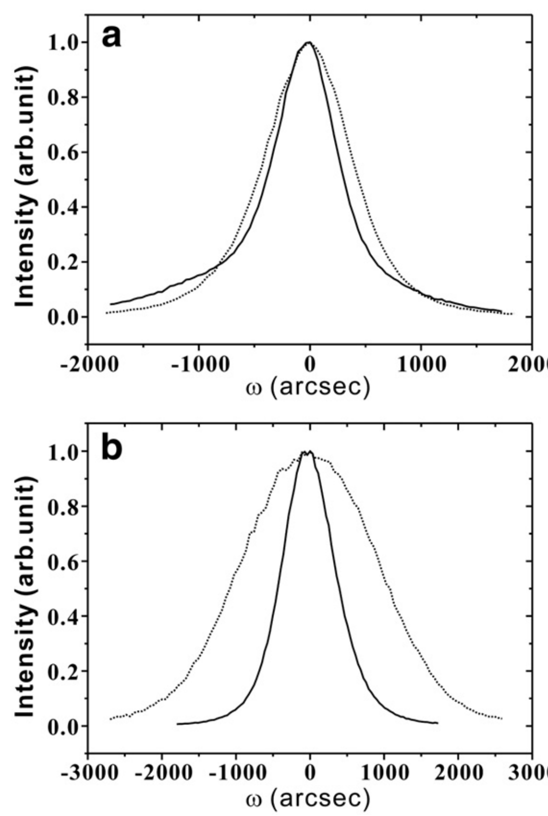

Fig. 3. (11$\bar{2}$0) X-ray rocking curves for the *a*-plane GaN films grown on conventional *r*-plane sapphire substrate (dash line), and on cone-shaped patterned *r*-plane sapphire substrate (solid line). (a) Scanning along the $[0001]_{GaN}$ direction and (b) scanning along the $[1\bar{1}00]_{GaN}$ direction.