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GaN-based light emitting diodes on nano-hole patterned sapphire substrate prepared by three-beam laser interference lithography*

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Nano-hole patterned sapphire substrates (NHPSSs) were successfully prepared using a low-cost and high-efficiency approach, which is the laser interference lithography (LIL) combined with reactive ion etching (RIE) and inductively coupled plasma (ICP) techniques. Gallium nitride (GaN)-based light emitting diode (LED) structure was grown on NHPSS by metal organic chemical vapor deposition (MOCVD). Photoluminescence (PL) measurement was conducted to compare the luminescence efficiency of the GaN-based LED structure grown on NHPSS (NHPSS-LED) and that on unpatterned sapphire substrates (UPSS-LED). Electroluminescence (EL) measurement shows that the output power of NHPSS-LED is 2.3 times as high as that of UPSS-LED with an injection current of 150 mA. Both PL and EL results imply that NHPSS has an advantage in improving the crystalline quality of GaN epilayer and light extraction efficiency of LEDs at the same time.

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Gallium Nitride (GaN)-based light emitting diodes (LEDs) have been widely used in outdoor full-color displays, back lighting and solid state lighting. However, the light extraction efficiency of GaN-based LEDs keeps non-ideal due to the internal total reflection (refraction) of the light between GaN and silica gel or sapphire, which results in low external quantum efficiency (EQE). Many techniques have been introduced to improve the light extraction efficiency of GaN-based LED, including surface roughening[1], Bragg reflector[2], metal mirror reflecting layer[3], photonic crystal[4] and patterned sapphire substrate (PSS)[5]. PSS has been proved to be able to both improve the crystalline quality of GaN layer[6] and enhance the light extraction efficiency[7] of GaN-based LED. As a consequence, it has been widely used in the production of high efficiency GaN-based LEDs. At present, the production of PSS always adopts traditional lithography to reduce the cost. The prepared PSS with a feature size of 1.5 μm thus is called micro-patterned sapphire substrate (MPSS)[8]. Compared with MPSS, nano-patterned sapphire substrate (NPSS) has a higher structure density and a theoretically far better light extraction efficiency[9]. The nanoscale patterns were usually fabricated by e-beam lithography, laser interference lithography (LIL)[10], nanosphere lithography[11] and nanoimprint lithography (NIL)[12]. However, it is difficult for e-beam lithography, nanosphere lithography and NIL to define wafer-scale nano-scale patterned structures with low cost, high efficiency and low defect.

In the past several years, a lot of researches on the fabrication of nanostructures by LIL have been reported[10,13-15]. In these studies, only Ref.[15] obtained nano-pillar patterned sapphire substrates by two-beam LIL. However, the improvement of the crystal quality of GaN epitaxial layers and the photoelectric properties of LED chip on the NPSS were not mentioned. In this paper, nano-hole patterned sapphire substrate (NHPSS) was fabricated using three-beam LIL system and dry etching technology, and GaN-based LED structure was grown on it. The structural, electrical, and optical properties of the LEDs grown on NHPSS and unpat-

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terned sapphire substrates (UPSS) are discussed.

Fig.1(a) shows the schematic diagram of the three-beam LIL system[13]. Diffraction grating with hexagonal patterns as shown in the right inset of Fig.1(a) was used to split the laser beam into three light beams. The three beams with equal intensity were positioned symmetrically around the central axis[16] with the same conical half-angle of θ by objective lens as illustrated in Fig.1(b). The numerical aperture (*NA*), work distance and field of view of the objective lens are 0.5, 5 mm and 80 μm, respectively. After step by step exposure, the surface of sapphire substrate with diameter of 49 mm consists of many shooters, and the seam between adjacent shooters is 2 μm. The lattice constant of hexagonal structure as shown in Fig.1(c) inside all shooters satisfies

$$a=b=\frac{2\lambda}{3\sin(\theta)} . \tag{1}$$

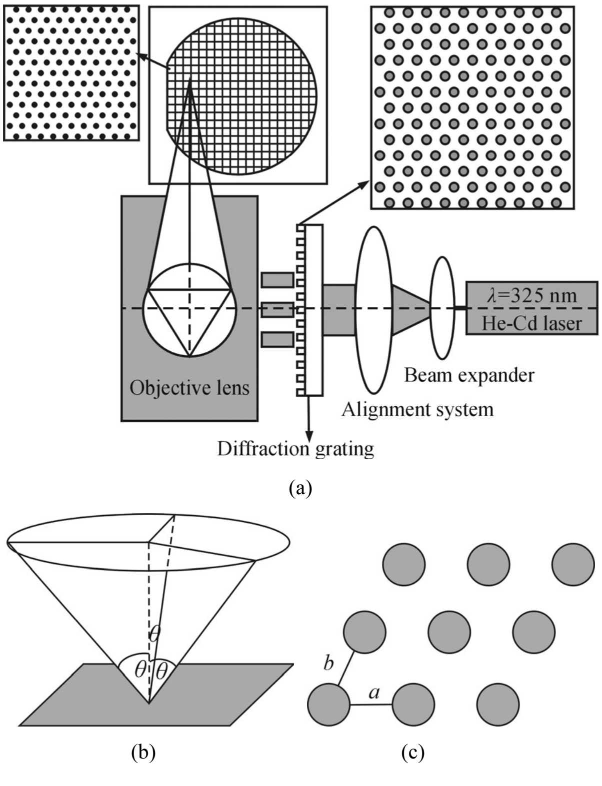
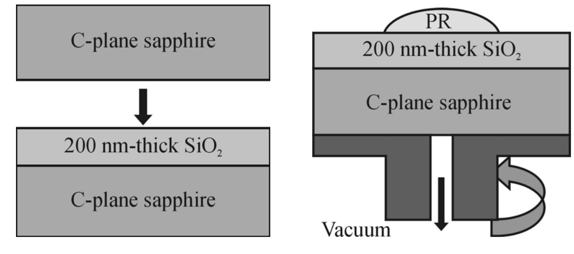


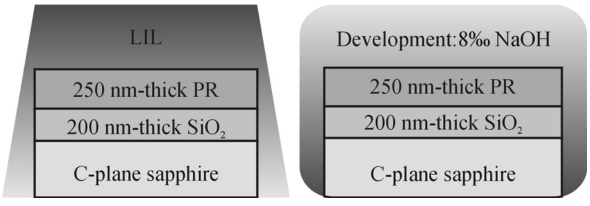
Fig.1 (a) Three-beam LIL system (The insets show the nano-hole pattern on PR(left) and the front of diffraction grating(right).); (b) Resulting wave vectors; (c) The lattice formed by the three-beam LIL system

NHPSS were fabricated by combining three-beam laser interference system with dry etching technique. Fig.2 shows the complete manufacturing process of NHPSS. Firstly, a 250 nm-thick SiO_2 layer was deposited on the sapphire wafer with diameter of 49 mm by using inductively coupled plasma chemical vapor deposition (ICPCVD). Secondly, a 250 nm-thick photoresist (PR) layer was spin-coated on the SiO_2 layer. After the exposure and development process, nano-hole patterns with the period of 550 nm and the diameter of 250 nm were fabricated on the PR. Then the nano-hole patterns were transferred from PR onto the SiO_2 layer by reactive-ion etching (RIE). In order to transfer nano-hole patterns onto the sapphire, dry etching process was executed using inductively coupled plasma (ICP) equipment[17]. Finally, the residual SiO_2 was removed by buffer oxide etcher (BOE), and NHPSSs were prepared. Atomic force microscope (AFM) image of NHPSS with size of 5 μm×5 μm is shown in Fig.3(a), and the thickness of the NHPSS along its one side is shown in Fig.3(b). It can be seen from Fig.3 that the period (*a*), hole radius (*r*) and hole depth (*h*) of the NHPSS are 550 nm, 180 nm and 100 nm (Fig.3), respectively.

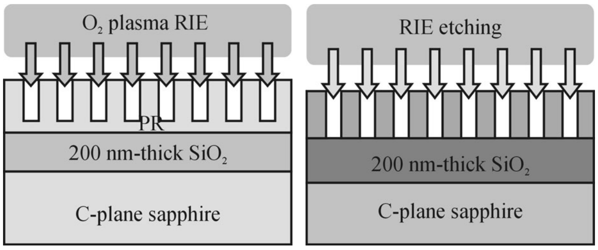
LED structure was grown on the NHPSS and UPSS using metal organic chemical vapor deposition (MOCVD) system[18]. The blue LED structure consists of a 30 nm-thick GaN nucleation layer grown at 520 °C, a 3 μm-thick un-doped GaN (UN-GaN) buffer layer grown at 1 070 °C, a 3 μm-thick Si-doped GaN (N-GaN) layer grown at 1 040 °C, an unintentionally doped InGaN/GaN multiple quantum well (MQW) active region with emitting wavelength of 460 nm grown at 770 °C, a 40 nm-thick Mg-doped p-GaN layer and a 8 nm-thick Mg-doped p-GaN contact layer grown at 1 050 °C. LED devices with chip size of 577 μm× 325 μm were fabricated by standard LED chip processes. An indium-tin-oxide (ITO) thin film with a thickness of 120 nm was deposited onto p-GaN surface. The n-contact and p-contact metal are Cr/Pt/Au layers with thicknesses of 30 nm/50 nm/1 400 nm.



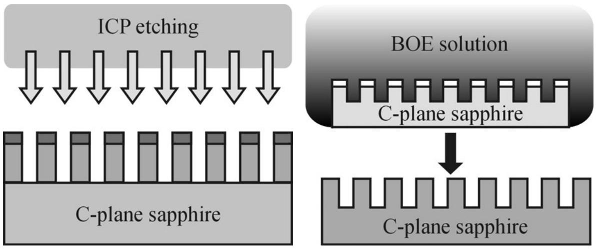
(a) Depositing SiO_2 by ICPCVD (b) Spinning photoresist



(c) Exposure by LIL (d) Development in 8‰ NaOH



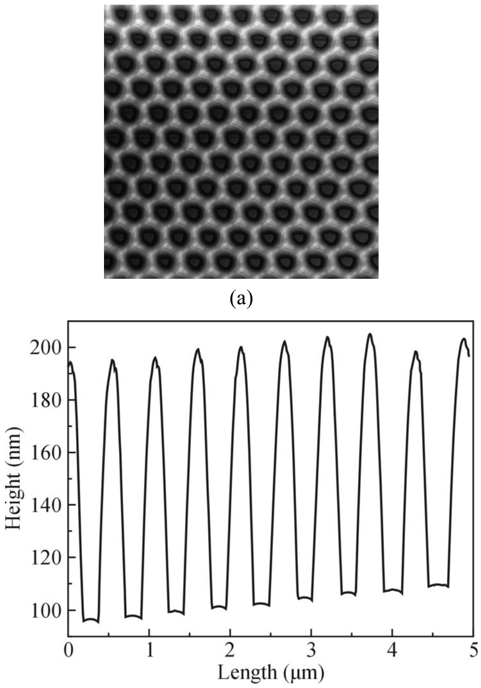
(e) Removing residual PR by RIE (f) Etching SiO_2 by RIE



(g) ICP etching of sapphire (h) Removing residual SiO_2 on NHPSS by BOE

Fig.2 Schematic diagram of the overall manufacturing processes of NHPSS

The crystal quality of the GaN layer was characterized by using high-resolution X-ray diffraction (HRXRD). According to the X-ray rocking curves (XRCs) of both (002) and (102) planes as shown in Fig.4, the full width at half maximum (*FWHM*) of (002) and (102) peaks of GaN film grown on NHPSS are determined to be 272" and 284" while corresponding values of GaN grown on UPSS are 293" and 318", respectively. Considering the two kinds of wafers were grown in the same process, the XRD results demonstrate that the GaN grown on NHPSS has a higher crystalline quality than that grown on UPSS.

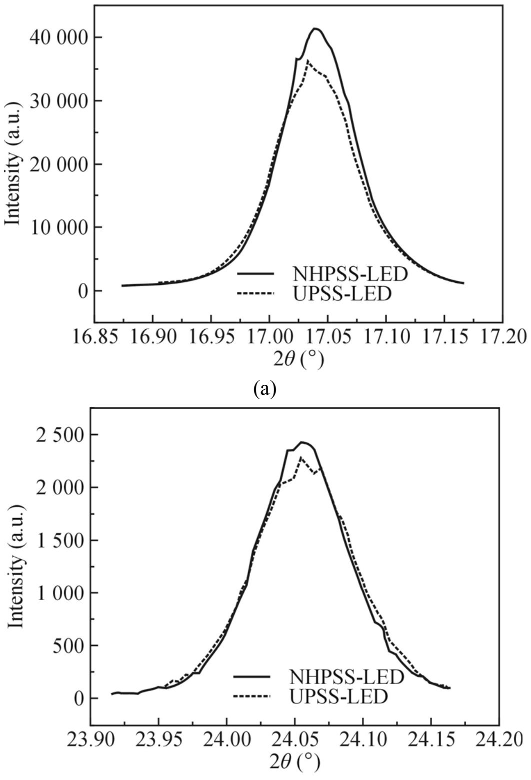


(a)

(b)

Fig.3 (a) AFM image and (b) thickness of NHPSS with size of 5 μm×5 μm

Photoluminescence (PL) measurements were conducted at room temperature with a 405 nm He-Cd laser as exciting source, and luminescence efficiencies of the two kinds of wafers were compared. As shown in Fig.5, the intensity of PL emission from NHPSS-LED is approximately three times stronger than that from UPSS-LED. *FWHMs* of the PL spectra for NHPSS-LED and UPSS-LED are 25 nm and 30 nm, respectively. The NHPSS-LED exhibits a 5 nm blue shift, which is caused partially by the residual strain released by the nano-hole patterns[19]. This result suggests that the nano-hole patterned structure can effectively improve the luminescence efficiency, in addition to its advantage in improving crystalline quality of GaN.



(a)

(b)

Fig.4 The XRCs of the (a) symmetric (002) planes and (b) asymmetric (102) planes for the LEDs grown on UPSS and NHPSS

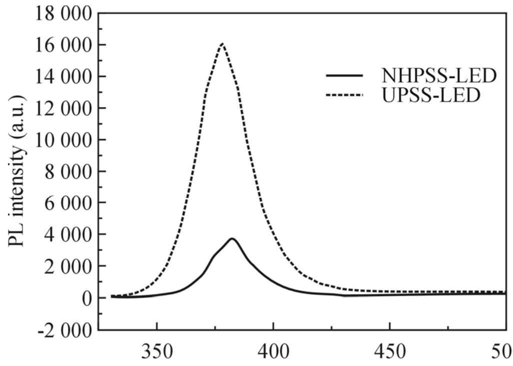


Fig.5 PL spectra of NHPSS-LED and UPSS-LED

Tab.1 lists the electroluminescence (EL) properties of UPSS-LED and NHPSS-LED. It is found that the reverse current (I_r) of UPSS-LED and NHPSS-LED at the reverse voltage of 8 V are 0.09 μA and 0.016 μA, respectively. The smaller leakage current of NHPSS-LED suggests that NHPSS can help to reduce the dislocation density of epitaxial films and to depress the leakage current consequentially. As shown in Tab.1, at the same injection current of 150 mA, the output power of NHPSS-LED is 81 mW, while that of UPSS-LED is only 36 mW. The luminescence efficiency of NHPSS-LED is enhanced obviously, which is 2.3 times as high as that of UPSS-LED. The luminescence efficiency enhancement of NHPSS-LED can be attributed to both the higher crystal quality of epilayers and the raised scattering effect of the nano-hole patterns.

Tab.1 EL property comparison of the UPSS-LED and the NHPSS-LED at the same injection current of 150 mA

LEDs	V_F (V)	I_r at −8 V (μA)	Optical power (mW)	Peak wavelength (nm)
UPSS-LED	3.334	0.090	36	474
NHPSS-LED	3.327	0.016	81	463

NHPSSs were successfully fabricated by a combination of low-cost and high-efficiency three-beam LIL and the dry etching technique. In the dry etching process, we can achieve different duty ratios (*r*/*a*) by choosing various gas proportion and etching bias. GaN-based LEDs were grown on NHPSS, and the HRXRD and PL results of the LED wafers imply that the crystalline quality of the epitaxial films on NHPSS is higher than that of films on UPSS. In addition, the PL results infer that nano-hole patterns can increase the scattering and then improve external quantum efficiency of the NHPSS-LED. The reverse currents of the UPSS-LED and NHPSS-LED are 0.09 μA and 0.016 μA at a reverse voltage of 8 V, respectively. The output power of LED grown on NHPSS is 2.3 times as higher as that of LED grown on UPSS at the same injection current of 150 mA. Both PL and EL results show that the nano-hole patterns can effectively improve the external quantum efficiency of NHPSS-LED. The effect of period and duty ratio on external quantum efficiency will be further investigated.

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