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Enhanced localisation effect and reduced quantum-confined Stark effect of carriers in InGaN/GaN multiple quantum wells embedded in nanopillars

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ABSTRACT

A conventional planar InGaN/GaN multiple quantum well (MQW)-based light emitting diode (LED) producing a blue-green dual-wavelength spectrum, due to phase separation in the InGaN well layers, is grown, and fabricated into a nanopillar (NP) LED array structure with an etching depth penetrating through the MQW region using a top-down dry etching process. Excitation power and temperature dependences of the photoluminescence (PL) spectra of the planar and NP samples have shown that the fabrication of the nanotexture not only decreases the quantum-confined Stark effect (QCSE), but also enhances the carrier localisation effect, both in the InGaN matrix region and in the In-rich quasi-quantum dot (QD) region, due to the reduction of the strain relaxation-induced piezoelectric field in the MQWs embedded in the nanopillars. Moreover, the fabrication of the nanotexture also improves the light extraction efficiency (LEE), due to the increased light-extracting surface area and light-guiding effect. These results are also consistent with the measured quantum efficiencies of the two samples.

1. Introduction

InGaN/GaN multiple quantum well (MQW) structures, as active layers, have been widely used in light emitting diodes (LEDs) due to the tenability of the emitted light from UV through visible spectral range [1–3]; however, due to the large discrepancy in atomic size between indium and gallium, and a large lattice mismatch of 11% between InN and GaN, either a phase separation or a slight composition fluctuation always occurs in the InGaN well layers [4–7]. In such a structure, on one hand, these In-rich regions (*i.e.*, so-called quasi-quantum dots (QDs)) act as carrier localisation centres, providing deep potential levels to suppress the outflow of carriers toward surrounding non-radiative centres [5,7–9], on the other hand, it also results in generation of structural defects that act as non-radiative recombination centres which is detrimental to the radiative recombination efficiency [5,7,10,11]. Also, due to a large lattice and thermal mismatch between the InGaN well layer and the GaN barrier layer, a large strain is induced in the MQWs, which can generate a large piezoelectric field in the MQW region. This induces a quantum-confined Stark effect (QCSE) therein, and therefore reduces the radiative recombination efficiency of the carriers in the MQWs [5,10,12]. Besides, as most commercial LEDs are fabricated on the basis of a conventional structure with a planar light-extracting surface, their light extraction efficiency (LEE) is limited by an internal reflection effect due to the large difference in the refractive index between the GaN ($n = 2.5$) and air ($n = 1$) [13,14]. Therefore, to obtain a high-quantum efficiency, the conventional *c*-plane InGaN/GaN MQW-based LED structures, especially for green LED structures with higher indium concentrations, should be optimised to improve the radiative recombination efficiency (*i.e.*, internal quantum efficiency IQE) and the LEE.

Recently, it has been reported that InGaN/GaN MQW-based nanopillar (NP) or nanowire (NW) LED array structures can effectively alleviate the aforementioned problems associated with conventional planar LEDs [15,16]: this is because the fabrication of the nanotextures penetrating through the MQW region can increase the free surface areas and surface/volume ratios which not only results in the reduction of the QCSE due to the relaxation of the strain in the MQWs regions embedded in these NPs or NWs, but also improves the probability of escape of photons inside the LED and light-guiding effect [17–22]. Therefore, the IQE and LEE can be significantly improved after the etching process

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[23,24]. However, it is noted that most of the related studies are mainly concerned with the effect of this nanotexture on the QCSE in the InGaN matrix and LEE of InGaN/GaN MQW LEDs with single wavelength emission arising from the InGaN matrix [15,16,23–25], in contrast, research into the effect of such nanotextures on the localisation effect of the carriers in the InGaN/GaN MQW LEDs producing a dual-wavelength emissions arising from the InGaN matrix and In-rich QDs due to a phase separation, is rarely reported, although it is important to obtain high-performance LEDs that emit different wavelengths with the appropriate power ratio.

In this study, a conventional planar InGaN/GaN MQW LED producing a blue-green dual-wavelength spectrum, due to phase separation in the InGaN well layers, was first grown on a (0001)-oriented sapphire using metal organic chemical vapour deposition (MOCVD). Then, based on the planar LED, an NP LED array structure with an etching depth penetrating through the MQW region was fabricated by using electron beam lithography (EBL) and inductively coupled plasma reactive ion etching (ICP-RIE). Microstructure properties of the planar sample were analyzed by high-resolution transmission electron microscopy (HRTEM), and the NP sample was structurally characterised using a scanning electron microscope (SEM). Finally, the excitation power and temperature dependences of the photoluminescence (PL) spectra from the two MQW structures are measured to clarify the underlying physics of carrier transfer and distribution as well as recombination therein.

2. Experiments

2.1. Planar LED fabrication

The InGaN/GaN MQWs-based planar LED (denoted by planar sample) was grown on a *c*-plane sapphire substrate using MOCVD. The precursors of Ga, In, N, and Si were trimethylgallium (TMGa), trimethylindium (TMIn), ammonia (NH_3), and silane (SiH_4), respectively. The epitaxial structure consisted of a 40-nm-thick low-temperature GaN nucleation layer, a 2-μm-thick unintentionally doped GaN buffer layer, a 2-μm-thick Si-doped *n*-GaN layer, and followed by 10 periods of InGaN (3 nm)/GaN (8 nm) MQWs as the active region, a 65-nm-thick low temperature *p*-GaN layer, a 28-nm-thick *p*-AlGaN layer, and a 200-nm-thick *p*-GaN layer. The structural sketch of the planar sample is shown in Fig. 1(a). The nominal indium content of the active region was approximately 32%.

2.2. Nanopillar array LED fabrication

An NP LED array structure (denoted as NP sample) was obtained starting from the aforementioned planar sample. Fig. 1 shows the process flowchart for the fabrication of the NP sample using a top-down etching method. First, a 60 nm-thick SiO_2 layer was deposited on the surface of the planar sample by plasma enhanced chemical vapour deposition (PECVD), then, a 200 nm-thick polymethyl methacrylate (PMMA) resist was spin-coated onto the sample (see Fig. 1(b)). The PMMA resist was then exposed by electron beam lithography (EBL; JBX-6300FS). The exposed PMMA resist was developed by methyl isobutyl ketone (MIBK): isopropyl alcohol (IPA) (1:3) at room temperature for 40 s, rinsed in IPA for 30 s and blew dry by pure nitrogen gas, and a uniformly distributed array of circular holes with a diameter of approximately 110 nm and pitch of approximately 150 nm was obtained (Fig. 1(c)). After developing, a 60 nm-thick Cr layer was deposited by e-beam evaporation (Peva-600E) (Fig. 1(d)). After lift-off in acetone, the patterned Cr nano-disk structure used as a mask for etching the SiO_2 layer by ICP-RIE (Oxford Instruments Plasmalab System100 ICP180), was formed on its surface. ICP-RIE operating at pressure of 10 mT with a C_4F_8/Ar flow of 10/40 sccm was used to etch the SiO_2 layer at a substrate temperature of 15 °C. ICP and RF powers were 300 and 30 W, respectively. The etching time was set to 30 s, corresponding to an etch depth of about 60 nm. As a result, the Cr/SiO_2 nano-disk structure, which was subsequently used as a mask for fabricating InGaN/GaN MQW NP arrays of the desired dimensions using ICP-RIE, was formed (Fig. 1(e)). For the ICP-RIE of InGaN/GaN active region, the flows of Cl_2, Ar, and BCl_3 were 18, 15, and 25 sccm respectively with an ICP/RF power of 300/50 W, an operating pressure of 10 mT, and at a constant temperature of 25 °C. The etching time was set to 4 min, corresponding to an etching depth of about 400 nm at which the etching depth is sufficient to penetrate through the entire active region (*i.e.*, reaching the Si-doped *n*-GaN layer, Fig. 1(f)). Subsequently, a Cr etchant ($(NH_4)_2Ce(NO_3)_6/CH_3COOH/H_2O$) and buffered oxide etchant (BOE: $HF/NH_4F/H_2O$) solution were used to remove the Cr and SiO_2 mask, respectively. At this point, the obtained NP LEDs are cone structures with oblique sidewalls. Finally, the InGaN/GaN MQW-based NP LEDs with smooth facet surfaces and vertical facet sidewalls were achieved by further etching in KOH-based etchant (AZ400K photoresist developer), as shown in Fig. 1(g).

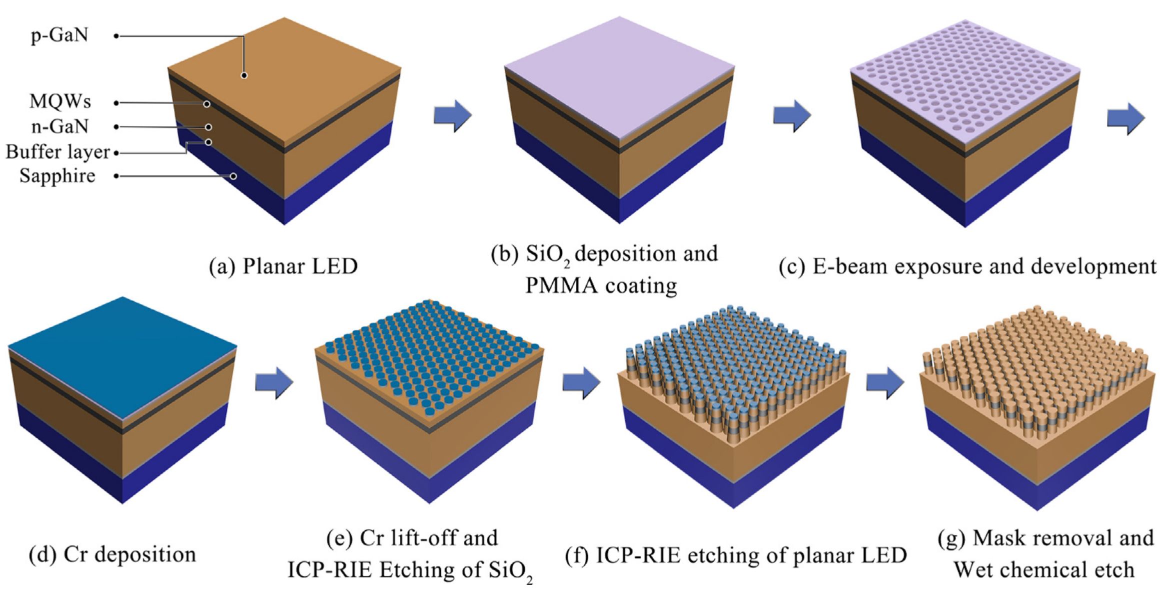

Fig. 1. Flowchart showing processing steps for the fabrication of InGaN/GaN MQW-based NP LED array structure using the top-down method.