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2005 Nanotechnology 16 448

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Nanotechnology **16** (2005) 448–450 doi:10.1088/0957-4484/16/4/020

Photoluminescence from $In_{0.3}Ga_{0.7}N$/GaN multiple-quantum-well nanorods

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Received 24 November 2004, in final form 12 January 2005
Published 11 February 2005
Online at stacks.iop.org/Nano/16/448

Abstract

The fabrication of $In_{0.3}Ga_{0.7}N$/GaN multiple-quantum-well nanorods with diameters of 60–100 nm and their optical characteristics performed by micro-photoluminescence measurements are presented. The nanorods were fabricated by inductively coupled plasma dry etching from a light-emitting diode wafer. The structure and surface properties of fabricated nanorods were verified by the field emission scanning electron microscopy and the transmission electron microscopy. The photoluminescence (PL) spectra with sharp linewidths of typically 1.5 nm were observed at 4 K. The excitation-power-dependent spectra show that no energy shift was observed for these sharp peaks. Moreover, increasing the excitation power instead leads to an occurrence of new, sharp PL peaks at the higher energy tail of the PL spectra, which suggest that excitons are strongly confined in quantum-dot-like regions or localization centres.

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1. Introduction

InGaN/GaN multiple quantum wells (MQWs) are utilized nowadays as the active layers of light-emitting devices based on wurtzite (hexagonal) group III nitrides and also have been attracting much attention as potential materials for short-wavelength light-emitting diodes (LEDs) because of the advantage of the tuning ability of the alloy bandgap. Many research groups have reported that the large compositional fluctuation is due to a large difference in either the lattice constants or the equilibrium vapour pressure of nitrogen between GaN and InN [1, 2]. It has been widely proposed that the large compositional fluctuations or phase separation of indium within InGaN/GaN MQWs may give rise to potential minima and QDs-like behaviour. In general, the depth of the trap centre was found to increase with the indium component. Moreover, spontaneous emission efficiency of InGaN/GaN MQWs has been recognized to be controlled by a balance between the exciton localization and wavefunction separation due to the quantum-confined Stark effect (QCSE), which result from electric field F normal to the QW plane owing to spontaneous and piezoelectric polarizations [3–5]. However, it has still not been confirmed that the photoluminescence (PL) spectra of InGaN/GaN MQWs consists of sharp and discrete lines. This is because quantum-confined structures in MQWs have slightly different sizes and compositions, or the period of compositional fluctuations is too short, leading to the observation of broad linewidth spectra. Therefore, the luminescence signals of individual potential minima cannot be observed easily. Recently, we have reported the fabrication of GaN-based nanorods by inductively coupled plasma (ICP) etching [6, 7]. In this paper, we present sharp PL peaks observed in $In_{0.3}Ga_{0.7}N$/GaN MQWs embedded within nanorods with a diameter of around 60 nm at 4 K.

2. Experiment

$In_{0.3}Ga_{0.7}N$/GaN MQW nanorods were fabricated from a conventional InGaN-based LED wafer grown by metallorganic chemical vapour deposition (MOCVD) on sapphire (0001) substrate. The LED structure consists of a 300 Å thick GaN nucleation layer, a 2 μm thick un-doped GaN, a 2 μm thick Si-doped n-type GaN, ten periods of $In_{0.3}Ga_{0.7}N$/GaN MQWs consisting of 6 nm thick $In_{0.3}Ga_{0.7}N$ well layers and 12 nm thick GaN barrier layers and a 30 nm thick Mg-doped $Al_{0.15}Ga_{0.85}N$ layer. Finally, a 250 nm thick Mg-doped

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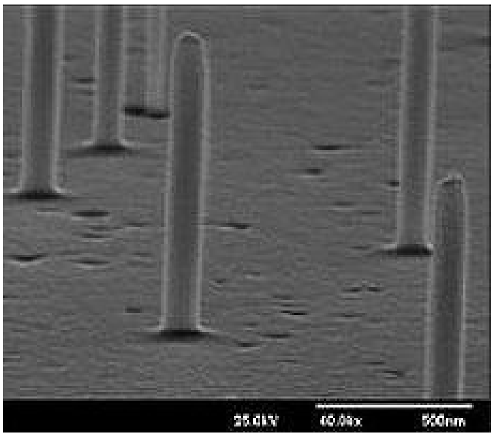

Figure 1. Scanning electron microscopy image of $\mathrm{In_{0.3}Ga_{0.7}N}$/GaN MQW nanorods.

p-type GaN contact layer was grown on the top region. The sample of grown wafer structure was subjected to the dry etching technique for nanorod formation using an ICP system (SAMCO RIE-101iPH). The etching process of the nanorods was performed under an inductively coupled plasma produced by a gaseous mixture of $\mathrm{Cl_2}$/Ar (10/25 sccm) at a chamber pressure of 20 mTorr. The ICP has a power of 200 W and a bias power of 200 W at an RF frequency of 13.5 MHz. The structure and surface properties of fabricated nanorods were measured by field emission scanning electron microscopy (FESEM), double-crystal high-resolution x-ray diffraction (HRXRD) and transmission electron microscopy (TEM). The optical properties of the nanorods were investigated by a micro-photoluminescence (μ-PL) set-up. For PL measurement, a doubled Ti:sapphire laser operating at 390 nm with a spot diameter of 40 μm and a liquid helium flow cryostat for low temperature were employed.

3. Results and discussion

Figure 1 displays a typical SEM image of $\mathrm{In_{0.3}Ga_{0.7}N}$/GaN MQW nanorods. The nanorods fabricated by ICP dry etching were almost vertical and of straight shape. The nanorods have lengths up to 500 nm and diameters ranging from 60 to 100 nm. Nanorods with diameters less than 55 nm were also observed. About 9–16 nanorods were probed in the μ-PL measurement based on the rod density of $\sim 3 \times 10^8$ estimated from figure 1. Because the nano-scale nanorods were distributed randomly, it is difficult to count exactly how many nanorods were probed in the μ-PL measurement. Structural characterization using TEM and HRXRD confirmed that the MQW structure in nanorods was intact in structure, as shown in figures 2(a) and (b) respectively. The thickness of the InGaN well and GaN barrier were estimated to be 6.2 and 12.2 nm which is in good agreement with the epitaxial design data.

A typical PL spectrum of InGaN/GaN nanorods under an excitation density of 0.9 W cm^{-2} was measured at 4 K as shown in figure 3. It consists of several discrete emission peaks whose positions are at 449, 453 and 457 nm respectively. The strong narrow emission peak at 457 nm has a full width at half maximum (FWHM) of about 1.5 nm. The position difference between each peak is estimated to be 4 nm (24 meV).

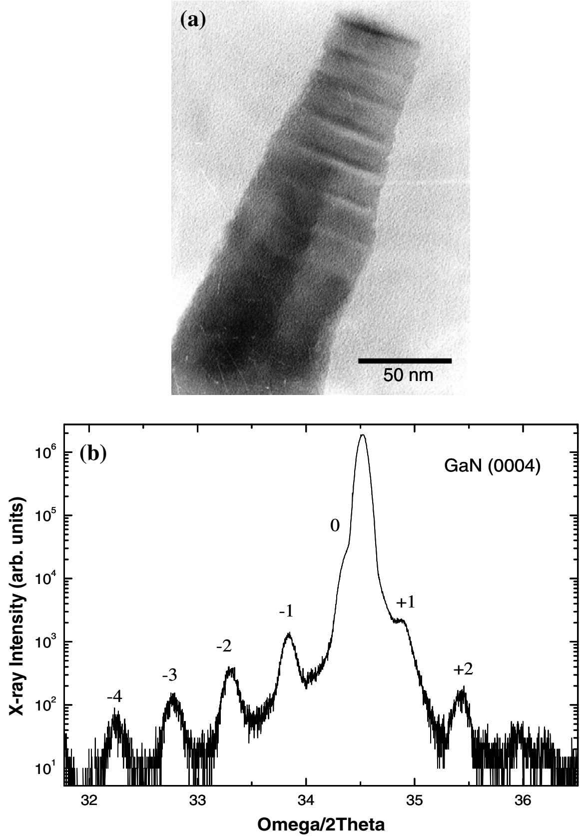

Figure 2. (a) Transmission electron micrograph of a single $\mathrm{In_{0.3}Ga_{0.7}N}$/GaN MQW nanorod. (b) Double-crystal x-ray diffraction spectra omega/2theta scans of the $\mathrm{In_{0.3}Ga_{0.7}N}$/GaN MQW nanorods.

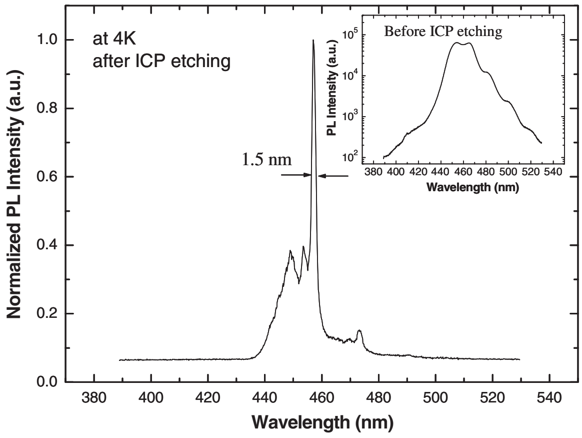

Figure 3. Photoluminescence spectrum of $\mathrm{In_{0.3}Ga_{0.7}N}$/GaN MQW nanorods excited under 0.9 W cm^{-2}. The inset is a photoluminescence spectrum of the as-grown bulk sample.

The inset in figure 3 is the spectrum from the as-grown bulk wafer before ICP etching, which was measured in the same condition for the nanorods. It shows a typical InGaN/GaN MQWs spectrum with a FWHM of about 26.5 nm and an undulation behaviour which is probably due to the Fabry–Perot interferences within the epitaxial layers [8]. Indeed, fabrication of nanorod structure from the $\mathrm{In_{0.3}Ga_{0.7}N}$/GaN MQWs bulk wafer does exactly show different behaviour from

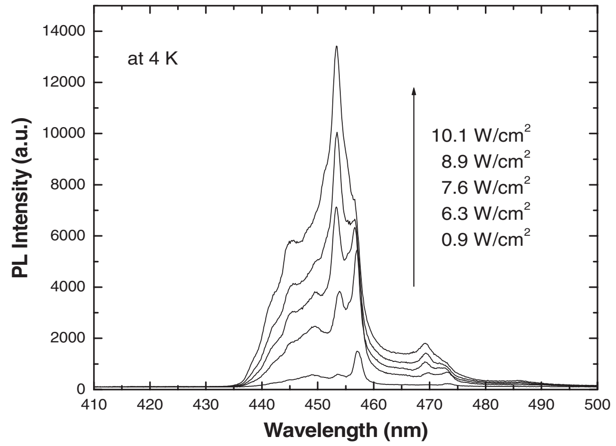

Figure 4. Excitation power dependent photoluminescence spectra of $In_{0.3}Ga_{0.7}N$/GaN MQW nanorods.

the typical PL emission spectra of bulk MQWs. This could be due to the decrease of inhomogeneous broadening in wells of nanorods.

An increase of the excitation density usually results in a pronounced and continuous blueshift of the inhomogeneously broadened PL spectra of InGaN/GaN MQWs, and there is still debatable discussion concerning the relative importance of bandtail filling and screening of internal electric field such as piezoelectric field induced by strain energy. The situation appears completely different, if one looks at the excitation power dependent PL spectra of InGaN/GaN MQWs embedded within small nanostructures. Figure 4 shows a series of spectra recorded at different excitation densities between 0.9 and 10.1 W cm^{-2} for $In_{0.3}Ga_{0.7}N$/GaN MQW nanorods at 4 K. Under low excitation densities, the e1–h1 peak at 457 nm is dominant. However, with increasing excitation density, the intensity of the peak on the high-energy side of the e1–h1 peak increases. Finally, this peak at 453 nm becomes dominant over the e1–h1 emission. The existence of three-dimensionally localized, QD-like states/structure has been demonstrated from the appearance of individual spectrally narrow emission lines and high-resolution transmission electron microscopy analysis in high indium composition ($x > 20\%$) InGaN/GaN MQWs [2, 9]. Thus, this is most likely a QD-like feature due to carrier localization and more efficient recombination through localized states [10]. On the other hand, the single exciton recombination peaks do not show any energy shift with increasing excitation power, demonstrating that no variation of the ground state energy due to screening by free carriers occupying delocalized states occurs. It also suggests that excitons are strongly confined in quantum-dot-like regions or localization centres.

4. Summary

In summary, we have successfully observed sharp PL peak emission from a QD-like structure embedded within $In_{0.3}Ga_{0.7}N$/GaN MQW nanorods fabricated by ICP dry etching using μ-PL measurement at 4 K. From the excitation power dependent PL results, we demonstrate the emission behaviour from single localization centres in the InGaN/GaN nanorods in this work. These experimental results suggest that excitons are strongly localized or confined in a QD-like structure. Such a circumstance presents interesting challenges to present efforts to develop blue nitride-based nano-optoelectronics devices, especially in the regime of higher indium concentrations, and must be taken into account for the design and fabrication of III–V nitride-based devices.

Acknowledgments

The authors wish to thank and express appreciation to Professor Y Yamamoto, Dr S Gotzinger, and H Deng of E L Ginzton Laboratory, Stanford University of California, USA, for their valuable discussions and technical support. This work is supported by the National Science Council of Taiwan under contract no NSC-93-2752-E-009-008-PAE and NSC-93-2120-M-009-006.

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