Solid-State Electronics 53 (2009) 336–340

Contents lists available at ScienceDirect

Solid-State Electronics

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

Optical properties studies in InGaN/GaN multiple-quantum well

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ARTICLE INFO

Article history:
Received 13 March 2008
Received in revised form 10 December 2008
Accepted 4 January 2009
Available online 27 February 2009

The review of this paper was arranged by Prof. E. Calleja

Keywords:
GaN
Light-emitting diodes
Localization effects
Photoluminescence
Longitudinal-optical-phonon

ABSTRACT

A comparative study on temperature-dependent photoluminescence (PL) of InGaN/GaN multiple-quantum-well (MQW) violet-blue light-emitting diodes (LEDs) is presented. For the violet-blue LEDs, the peak energy exhibits a well-established S-shaped temperature behavior. The redshift at low temperatures is explained by carrier relaxation into lower energy states, which leads to dominant radiative recombination occurring mostly at localized states. The temperature-dependent PL was attributed to the localization effects in the MQW region of the samples. Up to three phonon replicas were also observed in the side-band of the quantum well luminescence with an energy separation similar to the GaN longitudinal-optical (LO)-phonon energy (~91 meV). The properties of LO-phonon satellites were investigated as a function of the indium fraction and the well-width in a active layer at low temperatures.

1. Introduction

Group III-nitrides semiconductors have drew much attention in the past decade because of wide applications for the blue and ultraviolet optoelectronic devices such as detectors, light-emitting diodes (LEDs) and laser diodes (LDs) [1–3]. In contrast to conventional III–V semiconductors, the ternary alloy InGaN, because of its wide and alterable band gap, was widely applied in the blue and green LEDs, LDs and have achieved remarkable success in commerce [4,5]. InGaN-based LEDs show very high emission efficiency in spite of the existence of a high density of microstructural defects [6,7]. Therefore, the optical properties which depend on the characteristics of the wells, such as their width [8,11], number [9], GaN cap thickness [10], and InN fraction [12] has been widely studied, including an analysis of the phonon-assisted contribution. At present, it is of great importance to further understand the origins of the high quantum efficiency of the InGaN-based LEDs, in terms of the material physics, the optimizing practical device design and the growth of the materials. Previous studies showed that the localization effects resulting from spatially inhomogeneous indium distribution played an important role in the spontaneous emission of InGaN/GaN quantum-well structures. It was considered that the localized excitons in In-rich region acting as quantum dots were responsible for the high efficient radiative recombination. However, further investigations of recombination mechanism of the localized excitons in InGaN/GaN quantum wells are necessary. In this paper, we describe the study of the optical properties of the multiple-quantum-well blue and violet InGaN/GaN LEDs. we concentrate on the peak energy and the LO-phonon sidebands of the photoluminescence. The appearance of the LO-phonon sidebands (PSB) to the low energy side of main PL-peak, namely zero-phonon line (ZPL), indicated high crystal quality and the coupling of the excitons to phonons. And their relative strength indicated the degree of exciton localization and the spatial separation of the electron and hole wave functions. The relative strength of the LO-phonon sidebands can be expressed according to Huang-Rhys (S) factors. For InGaN/GaN heterostructures, the degree of exciton localization as well as the spatial separation of the electron and hole wave functions enhanced by the strong built-in electric fields are two critical factors that influence the LO-PSBs. Their relative contributions are hard to separate. Nevertheless, the properties of the PL are shown to provide some details.

2. Experiments

A series of blue-violet InGaN/GaN multi-quantum-wells (MQWs) LED samples were grown on (0001) sapphire (α-Al_2O_3) substrates by using low pressure metal-organic chemical-vapor-deposition (LP-MOCVD). Trimethylindium (TMI), trimethylgallium (TMG) and ammonia (NH_3) have been used as precursors of In,-Ga,and N sources, respectively. H_2 or N_2 were used as carrier gases. The device structures were identical, as follows: a low-temperature GaN buffer layer, a Si-doped n-GaN, a 5-period undoped-InGaN/

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0038-1101/$ - see front matter
doi:10.1016/j.sse.2009.01.005

GaN MQW layers, a p-type AlGaN cladding layer, and a p-GaN contact layer. The active layer of InGaN quantum well were grown at different temperatures between 730 °C and 780 °C, with the temperature varied in uniform steps of 10 °C, leading to six samples in all. The quantum well and barrier layers were grown in low temperatures due to the high saturated vapor pressure of InN.

The In compositions in InGaN layers were determined by the measurement of X-ray diffraction (XRD) curves using QCbeta200 diffractometer at wavelength is 1.54056 Å. The PL spectra were excited by 325 nm light of HeCd laser and detected by Si-detector after splitted by monochromator. The samples were cooled to ≈20 K using a closed cycle helium refrigerator. The optical characteristics of the samples were measured in the temperatures range 20–300 K.

3. Results and discussion

The LED samples have similar crystal characteristics which are revealed by the results of the XRD. Fig. 1 shows the representative double crystal XRD ω–2θ scanning curves of the (0002) crystal planes of InGaN/GaN LEDs samples whose InGaN layer were grown at 740 °C, 760 °C and 780 °C, respectively. There are 5–6 orders of diffraction satellites in the XRD spectrum in condition that the scan range is large enough, which indicates good crystalline and structure quality of the samples. The indium-mole fractions and the thickness of the quantum well are deduced from XRD diffraction spectra using the software Rads Mercury. The details for simulation are as follows: The number of wells is five with undoped GaN barriers of ~16 nm. The relaxation in every layer were handled as follows: substrate GaN layer 100%, well layer InGaN 0%, and barrier layer GaN 100%. As a result, the indium-mole fractions fall dramatically from 0.18 to 0.08 with the growth temperature increasing from 730 °C to 780 °C. This decreasing is attributed to the high saturated vapor pressure of InN. As the temperature increases, the indium desorption enhances. Ensuingly, the indium-mole fractions in the well layer decreases, and the thickness of the well reduces at the same time.

The temperature-dependent PL spectra of the two series LEDs are plotted in Fig. 2. There are several obvious features in this figure. First, for all samples, Owing to the weakened influence of nonradiative recombination, the MQW emission intensity increase rapidly with temperature reducing. Second, the MQW emission peak exhibits firstly a slight blueshift with temperature decreasing

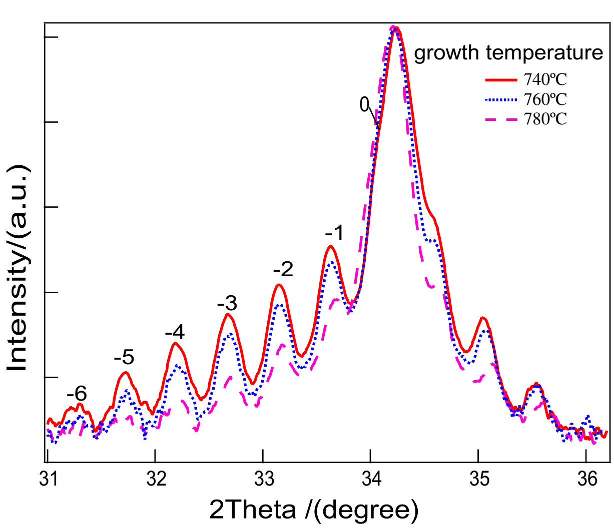

Fig. 1. The double crystal X-ray $\omega \sim 2\theta$ scan spectra of (0002) planes of InGaN/GaN MQW samples.

from room temperature to 200 K, then a pronounced redshift at lower temperatures and finally a large blueshift with temperature decreasing from 60 K to 20 K. In addition, the deep-level emission centered at 2.2 eV of the yellow band is suppressed at temperatures below 200 K, indicating a freezing out of defects in the active region. Finally, all samples have the similar emission spectra: they consist of a main feature followed by a series of LO-phonon replicas appearing at the low energy side of the main emission band. At low temperatures, the LO-phonon satellites peaks are clearly resolved in all samples. The energy separation of the LO-phonon satellites remains constant at ~91 meV which is close to the LO-phonon energy of GaN, as observed previously [8–12]. The LO-phonon replicas appear at different temperatures for different samples. The LO side bands occur below 100 K for violet LEDs, but at 200 K for blue LEDs. This indicated that the coupling of the excitons to phonons in the blue-LEDs is stronger than that in the violet-LEDs, namely the degree of the exciton localization and the spatial separation of the electron and hole wave functions in the blue-LEDs is much stronger than those in the violet-LEDs. This is because the blue-LEDs grown at lower temperatures have higher Indium fraction in the well layer than the violet-LEDs grown at higher temperatures. Fig. 3 shows the representative low-temperature PL spectrum measured from the "490 nm" InGaN/GaN MQW LED whose active layer is grown at 730 °C. There are two phonon satellites at the lower energy side of the PL spectra. As illustrated in this figure, The peak emission energy, FWHM of the ZPL and its phonon satellites are estimated using multi-Gaussian peak fitting following substraction of a linear background. The LO-phonon replicas which broadening the low energy side of the PL pectra are constrained to have the same FWHM as the ZPL.

Fig. 4 illustrates the temperature-induced shifts of peak-emission energy of the LEDs. When the temperature changes from 300 K to 60 K, both the peak energies of the violet and blue LEDs exhibit similar blueshift–redshift behavior. For the violet LEDs, the blue-shift from 300 K to 200 K is much pronounced than that of the blue LEDs, but the red-shift from 200 K to 60 K is much slight than that of the blue LEDs. The S-shape of the PL spectrum can be explained as follows: at low temperatures, the thermal energies of the carriers are not high enough to overcome the localization potential and the carriers are usually localized. The localized states in the InGaN layers are caused by potential fluctuations, because of inhomogeneous Indium content. For the blue LEDs, the fluctuations of the Indium content in the InGaN layer is even serious leading to the pronounced redshift between 200 K and 60 K. The carriers localized in the lower energy states may result in the change of the carrier recombination mechanism. At room temperature, band-to-band transitions may be dominant, whereas recombination at localized states becomes important at low temperatures. As temperature decreases further below 60 K, the peak energy change slighly in the violet LEDs, but shift to higher values in the blue LEDs. This difference reflects different carrier recombination mechanism in the violet LEDs and the blue LEDs in this temperature range. The PL peak shift in the violet and blue LEDs with an increase of temperatures above 200 K follows the typical temperature-dependent GaN energy-gap shrinkage.

Fig. 5 shows the Normalized low-temperature ($T \sim 20$ K) photoluminescence spectra of InGaN/GaN MQW samples with various indium fractions. All of the spectra have similar shape. The measured PL peak energy of the zero-phonon recombination (3.24 eV, 3.18 eV, 2.91 eV, 2.78 eV, 2.67 eV and 2.54 eV) decrease with an increase of indium content of the quantum wells from 0.08 to 0.18, which results from the growth temperature of InGaN layer. For example, when the indium content is 0.18, the emission energy is 2.54 eV. when the indium content decreases to 0.08, the emission energy increase to 3.24 eV accordingly. At the same time, the strength of the phonon replicas relative to the zero-phonon