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Cathodoluminescence study on in composition inhomogeneity of thick InGaN layer

H. Wang [a,*], D.S. Jiang [a], U. Jahn [b], J.J. Zhu [a], D.G. Zhao [a], Z.S. Liu [a], S.M. Zhang [a], H. Yang [c]

[a] State Key Laboratory on Integrated Optoelectronics, Institute of Semiconductors, Chinese Academy of Science, PO Box 912, Beijing 100083, China
[b] Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, 10117 Berlin, Germany
[c] Suzhou Institute of Nano-tech and Nano-bionics, Chinese Academy of Sciences, Suzhou 215123, China

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ABSTRACT

We have investigated the optical properties of thick InGaN film grown on GaN by cathodeluminescence (CL) spectroscopy. It is found that there is obvious In composition variation in both growth and lateral direction of InGaN film. The depth distribution of In composition is closely related to the strain relaxation process of InGaN film. Accompanied with the relaxation of compressive strain, the In composition of InGaN layer increases and the CL peak energy shifts towards red. Moreover, a rather apparent In composition fluctuation is found in the relaxed upper part of InGaN layer as confirmed by CL imaging.

1. Introduction

InGaN alloys cover the spectral range continuously from near-infrared (InN, 0.65 eV) to near-ultraviolet (GaN, 3.4 eV), which offers a potential to develop high-efficiency tandem solar cell based on nitride materials [1]. Differing from conventional InGaN-based optoelectronic devices such as light emitting diodes and laser diodes, in which fully strained InGaN thin layers (in order of several nanometers) are used in the active region, fabrication of InGaN/GaN solar cells requires a much thicker InGaN layers (larger than 200 nm) to maximize the absorption of incident light.

However, the growth of high-quality thick InGaN layers, especially for InGaN with relatively high In content, is very difficult. Due to the large lattice constant mismatch between InN and GaN (~11%), even a little higher In content in InGaN ternary alloy changes its lattice constant remarkably. As a result, with increasing layer thickness, InGaN layer is hard to keep coherent in lattice with underlying GaN template and the relaxation of misfit strain will lead to a degradation of InGaN crystalline quality. Moreover, as an important feature of ternary compounds, the composition inhomogeneity as a result of phase separation of InGaN has been frequently observed [2,3]. It is reported that In composition variation is closely related to the strain state in InGaN films [4]. Therefore, a comprehension of strain transformation and In composition variation in thick InGaN layer is very important for further improvement of epitaxial structure and device performances.

In this study, we have investigated the optical properties of InGaN film by spatially resolved cathodeluminescence (CL) spectroscopy. It has been found that there is obvious In composition variation both along growth direction and in the lateral direction. The In composition variation is proposed to be closely related to strain relaxation process of mismatch strain in InGaN film.

2. Experimental details

InGaN layers were grown in a horizontal metalorganic chemical vapor deposition (MOCVD) reactor. The 2 μm-thick GaN layers were firstly grown on c-plane sapphire substrates by MOCVD and served as pseudo-substrates for InGaN growth. Triethylgallium (TEGa), trimethylindium (TMIn) and ammonia were used as Ga, In, N precursors, respectively. Prior to InGaN deposition, the as-grown GaN templates were thermally cleaned at 700 °C in NH_3. Then, InGaN layers were grown at 720 °C under a nearly identical condition for all samples with a nominal thickness of 450 nm.

Low-temperature CL investigations on the samples were performed in a scanning electron microscope (SEM) equipped with an Oxford mono-CL2 and He-cooled stage operating at 6 K. A grating monochromator and the cooled charge-coupled device array/photomultiplier were used to disperse and detect the CL signal, respectively. A Bruker D8 Discover X-ray diffractometer with a monochromatic Cu Kα1 X-ray was used to perform X-ray diffraction (XRD) reciprocal space mapping (RSM) measurements.

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* Corresponding author. State Key Laboratory on Integrated Optoelectronics, Institute of Semiconductors, Chinese Academy of Science, PO Box 912, Beijing 100083, China. Tel.: +86 10 82304168; fax: +186 10 82304242.
E-mail address: wangh@semi.ac.cn (H. Wang).

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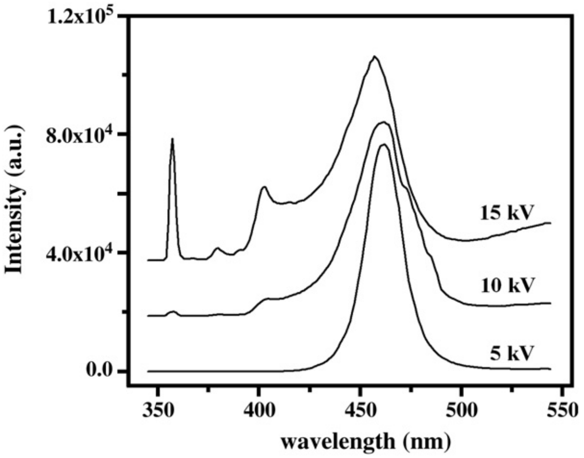

Fig. 1. CL spectra of an InGaN/GaN sample measured at different electron acceleration voltages of 5, 10, and 15 kV, respectively. The sample temperature was 6 K.

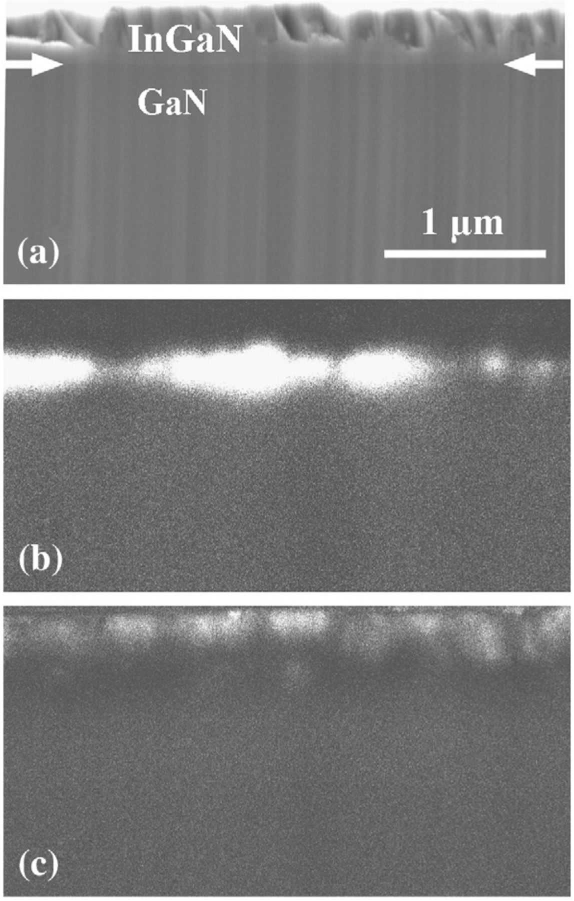

Fig. 2. Cross-sectional images from the same area of the InGaN/GaN sample (a) SEM image, (b) CL image at $\lambda = 402$ nm, and (c) CL image at $\lambda = 461$ nm measured at an electron acceleration voltages of 5 kV. The white arrows indicate the position of InGaN/GaN interface.

3. Results and discussion

Fig. 1 shows the CL spectra measured from the top surface of the InGaN/GaN sample for a varying electron acceleration voltage of the electron beam at 6 K. For an acceleration voltage of 5 kV, only one InGaN CL peak at the wavelength of 461 nm is observed. When the acceleration voltage increases, more peaks appear in the CL spectra. For the acceleration voltage higher than 10 kV, one starts to observe another InGaN related emission at 402 nm and the emission peaks from GaN layer, i.e., the near band emission of GaN at 357 nm, and donor–acceptor pair related emission at 379 nm. When the acceleration voltage increases from 10 to 15 kV, both GaN near band gap emission and InGaN related emission at 402 nm were enhanced. It is known that the electron penetration depth increases with increasing acceleration voltage. Therefore, the electron excited region extends progressively from near surface region to the InGaN/GaN interface with increasing voltage [5]. This fact also indicates that the InGaN related emission peak at 402 nm should originate from the region far from the InGaN top surface.

Cross-sectional CL imaging has been carried out to investigate the origin of double CL peaks from the InGaN film. Fig. 2(a) shows the cross-sectional SEM image of InGaN/GaN sample cross-section measured by the second electron imaging. It can be seen that there are some pits on the InGaN surface and many grooves on InGaN cross-section caused by the sample cleavage. Corresponding to the wavelength of the two peaks in Fig. 1, we have recorded two cross-sectional CL images for the wavelength of 402 and 461 nm in Fig. 2(b) and (c), respectively. The results clearly confirm that the high-energy peak (402 nm) of the InGaN CL spectrum originates from the near InGaN/GaN interface region, while the top part of InGaN layer exhibits obviously a longer wavelength emission at 461 nm. It means that the CL peak energy shifts towards red with increasing InGaN thickness. It is known that both relaxation of compressive mismatch strain and increase of In composition in InGaN alloy can cause a red shift of CL peak. However, it is noted that the observed large red shift of InGaN peak wavelength with increasing thickness (as large as 58 nm) is so large that cannot be accounted for only by the strain relaxation. There should be an increasing InN mole fraction distributed along the growth direction.

To understand better about the evolution of the strain relaxation and In composition variation with increasing InGaN layer thickness, an XRD RSM of the InGaN/GaN sample is recorded around $(10\bar{1}5)$ reflection. As shown in Fig. 3, the two vertical and inclined dashed lines in the RSM mark the locations where the reciprocal lattice points (RLPs) of pseudomorphic and completely strain–relaxed films would appear, respectively [6]. The solid lines in Fig. 3 are isocomposition lines which connect the pseudomorphic and completely relaxed lines, indicating the calculated relaxed directions for two In compositions of

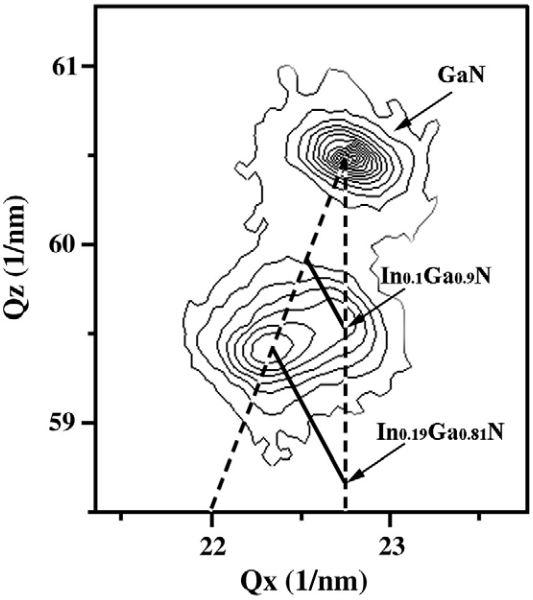

Fig. 3. Reciprocal space mapping of X-ray diffraction intensity from the $(10\bar{1}5)$ reflection for the two InGaN/GaN samples.