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Investigation of alloy Composition Fluctuation in Indium Gallium Nitride (InGaN) Tenary Films

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Abstract:
In a bid to reduce the rate of nonradiative recombination in InGaN LEDs, the optical and structural properties of the ternary films were investigated by temperature-dependent photoluminescence (PL), high-resolution X-ray diffraction (HRXRD) and high-resolution electron microscopy (HREM). The results show that the temperature-dependent PL intensity of the InGaN film is similar to that of the disordered alloys, which is thought to be due to local alloy compositional fluctuations (ACF) in the epilayer. HRXRD measurement reveals that, there are In-rich and In-poor phases in the film and HREM observation, on the other hand, demonstrates that nanoclusters formed in the epilayer. Therefore the experimental results support the existence of ACF in the epilayers which causes nonradiative recombination.

Keywords: *Photoluminescence, high-resolution X-ray diffraction, alloy compositional fluctuation.*

1. Introduction

Group-III nitrides, GaN, InN and their alloys, have been extensively studied in recent years because of their applications for optoelectronic devices operating in the range from blue-green to ultraviolet [6]. The ternary InGaN alloys have attracted much attention as active-layer materials for fabrication of short-wavelength light-emitting diodes (LEDs) and laser diodes (LDs) [6, 8]. However, lattice mismatch between the InGaN layer and the barrier layer (GaN) causes built-in internal electric fields that influence the emission intensity or quantum efficiency (QE) [2]. Recently, $In_xGa_{1-x}N$ alloys have been regarded as the most promising candidates to replace AlGaN or GaN as barriers due to their bandgap and lattice constant could be independently adjusted by varying the Indium compositions. Consequently, lattice-matched quantum wells (QWs) or hetero structures under the required bandgap energy can be obtained [1, 4]. High-quality $In_xGa_{1-x}N$ alloys, that can be successfully used as the barrier or active layer of high QE LEDs, grown using metal organic chemical vapor deposition (MOCVD) is available today in the consumer market [1,7]. However, since the composition fluctuation is an important phenomenon in InGaN, more investigations are required on the ternary in this regard. As a consequence, the objective of this work is to investigate the compositional fluctuation in the InGaN epitaxial film grown by MOVPE, by measurement of the optical and structural properties using temperature-dependent PL, HRXRD and HREM techniques.

2. Materials and Methods

The InGaN films were subjected to investigation with detailed summary as follows: The cross-section specimens for HREM observations were prepared in the manner explained by [3], by cutting, gluing, mechanical polishing, dimpling and Ar^+-ion milling on a liquid nitrogen-cooled holder in order to minimize ion beam damage. The thickness of the ternary epilayers were measured by transmission electron microscope (TEM) and the microstructure investigations were carried out on a CM200-FEGTEM with a point resolution of 0.23 nm and line resolution of 0.1 nm. Energy dispersive X-ray analysis (EDX), calibrated firstly according to the method by [4], was performed to measure the chemical compositions of the films. For PL measurements, the samples were excited by a He-Cd laser working at 325 nm; the spectra were detected by a cooled GaAs photomultiplier tube. HRXRD, using synchrotron radiation with a wavelength 0.1540 nm as a light source, was applied to investigate the structural properties.

3. Results

The thickness of the ternary films is 0.6 μm as measured by TEM. Table 1 lists the growth parameters and the composition of the ternary films. Figure 1 shows PL spectra of the epitaxial samples taken at 10 K. The PL spectra show the broad bands peaked from 3.00 eV to 3.32 eV and their full width at half-maximum (FWHM) decrease from 172 meV to 140 meV with the decrease of Indium incorporation. Therefore, Indium content in the ternary films was a main factor that influences the FWHM and it is probable that the existence of large ACF broadens the FWHM. Figure 2 shows the integrated PL intensity of Sample L008 versus temperature together with the lifetimes at the emission peak versus temperature. It was found that the temperature dependence of the PL intensity can be well fitted by the following equation [7, 9],

$I_{PL} = I_0/ [1+ an \exp (T/T_0)]$ ……………… (1)

Where I_{PL} is the PL intensity, T is the absolute temperature and T_0 is the characteristic temperature corresponding to the energy depth from the radiative state to nonradiative state, A is the tunneling factor and I_0 is the luminescence intensity at low-temperature limit.

Figure 3 shows a high-resolution image of the In0.1Ga0.78N layer, in which the non-homogeneous nanoclusters are indicated. The nanoclusters display different darker contrast from the matrix because of the different alloy compositions between them. A more detailed observation of the HREM image reveals that the lattice inside the darker region is slightly expanded in comparison to that in the surrounding matrix, implying a larger lattice constant of the darker region. The lattice expansion is thought to be produced by a higher indium concentration at that region. Series of measurements show that the lattice constants are 0.5223 nm and 0.5209 nm corresponding to the darker region and the surrounding matrix, respectively. The values are consistent with those of HRXRD measurement. Therefore the results demonstrate the existence of the alloy compositional fluctuations in the InGaN epilayer.

Sample No.	**L008**	**L027**	**L006**	**L009**
Growth Temp. (◦C)	820	830	850	870
In molar content	0.10	0.06	0.029	0.033

Table 1: the growth conditions and alloy composition of the InGaN Epilayers

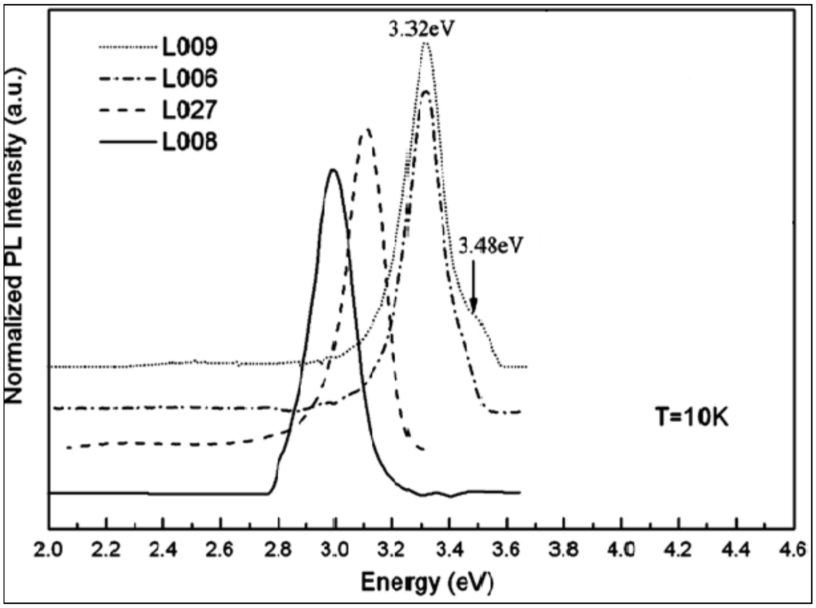

Figure 1: Low-temperature (10 K) PL spectra of the InGaN epilayers grown at different temperatures.

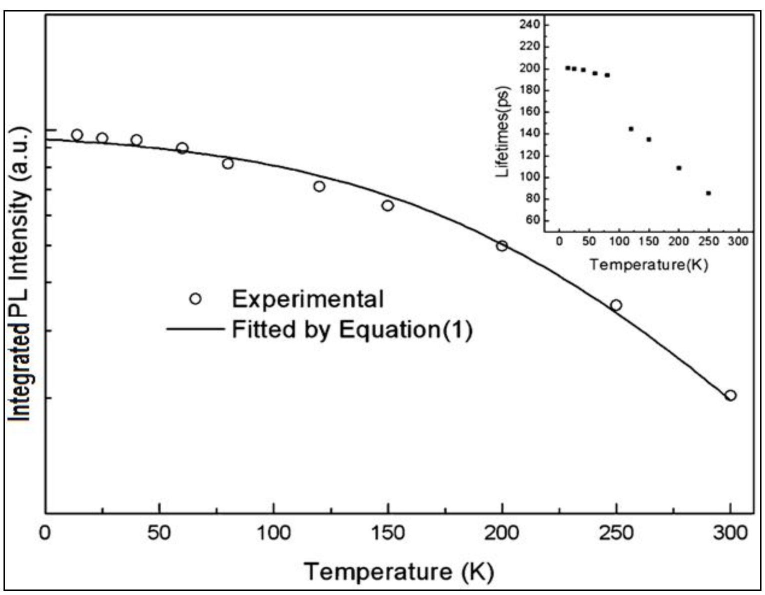

Figure 2: The dependence of integrated PL intensity for the In0.1Ga0.78N epitaxial film on temperature. The temperature-dependent PL intensity shows a relationship of $I_{PL} \propto [1+ A \exp (T/T_0)]^{-1}$. The inset is the temperature-dependent lifetime

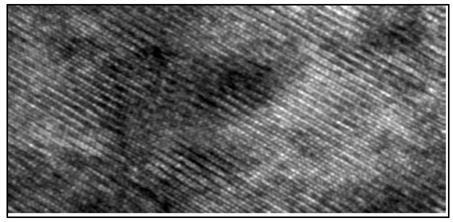

Figure 3: A cross-sectional HREM image of the In0.1Ga0.78N epilayer

5. Discussion

The lifetime changes little when the temperature is below 80 K, indicating that radiative recombination dominates the emission below 80 K. However, above 80 K, the lifetime decreases rapidly, therefore, there appears to be nonradiative recombination. In this context, it is inferred that T_0 must be less than 80 K, which, in fact, is calculated to be 73.5K comparable with the T_0 detailed by [5]. Also, equation 1 is valid in the case of existence of localized band-tail states [9]. The existence of localized excitons has often been suggested in the InGaN films and QW structures because of the Indium fluctuation [6]. Therefore, it seems reasonable to infer that the PL signal in the InGaN epilayer comes from the localized states. These states can be attributed to local compositional fluctuation in the InGaN film. In fact, the InGaN material system could be regarded as a pseudo-ternary system because mixing of Indium and Gallium atoms occur on one of the sub lattices. Moreover there exists strong immiscibility between the InN and GaN, so it is expected that the InN-like or GaN-like nanoscale clusters can be easily formed. These clusters become the localized center of excitons (carriers).

6. Conclusions and Recommendation

In summary, the temperature-dependent PL, HRXRD and HREM have been used to investigate the alloy properties of the InGaN alloy epilayers. The temperature dependent PL result showed that the ternary alloy exhibits the characteristics of disordered semiconductors, the reason for this the epilayer and HREM observation revealed the formation of the nanoscale clusters in the film. Therefore, the above results demonstrated the existence of ACF in the ternary. To achieve is supposed to the existence of ACF in the film. The HRXRD measurement suggests the existence of high indium and low indium InGaN in better radiative recombination, the variation in Indium composition should be as minimal as possible while the temperature should be kept below 80 K.

7. References

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