Effect of Barrier Temperature on Photoelectric Properties of GaN-Based Yellow LEDs *

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The effect of growth temperature of barriers on photoelectric properties of GaN-based yellow light emitting diodes (LEDs) is investigated. It is found that as the barrier temperature increases, the crystal quality of multi-quantum wells (MQWs) and the quality of well/barrier interface are improved, and the quantum well is thermally annealed, so that the indium atoms in the quantum well migrate to the equilibrium position, reducing the phase separation of the quantum well and improving the crystal quality of quantum wells (QWs). However, the external quantum efficiency (EQE) of the samples begins to decrease when raising the barrier temperature even further. One explanation may be that the higher barrier temperature destroys the local state in the quantum well and reduces the well/barrier interface quality. Therefore, a suitable barrier temperature is proposed, contributing to the improvement of the luminous efficiency of the yellow LEDs.

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Since the pioneer work on visible-spectrum LEDs in 1962,[1] great achievements have been made in solid state lighting in the past half century. LEDs in the entire visible light range have been successfully developed, providing us with a colorful artificial vision world. However, multi-primary full-spectrum LEDs are not maturely developed. Blue and red LEDs are very efficient with external quantum efficiency (EQE) over 80% and 50%,[2,3] respectively, whereas yellow LEDs are inefficient.

When the wavelength of an AlGaInP system changes from red to yellow, the band gap transits from direct to indirect, thus the possibility of obtaining high luminous efficiency in the yellow light band is restricted. Actually, the challenge in InGaN systems is the material growth. Usually, the relatively lower efficiency for long wavelength of InGaN-based LEDs can be mainly attributed to high indium content. To obtain high indium composition of InGaN, a lower temperature is required in growth process of InGaN quantum wells, which restrains the cracking efficiency of ammonia and reduces the diffusion ability of the atoms, resulting in coarse interface, high dislocation density and high point defect density.[3,4] Moreover, more indium would enhance indium phase separation owing to the lower miscibility of InN in GaN. These also seriously affect the luminous efficiency of yellow LEDs.

To achieve high-quality InGaN quantum wells, the effects of barrier growth conditions on the GaN-based LEDs were also investigated prior to preparation, especially the short wavelength.[5–8] The presence of H_2 contributes to increase of growth rate of barriers and to decreases of both the indium content and the thickness of QWs.[9] Increase in growth temperature of quantum barriers of LEDs can effectively improve crystal quality of GaN quantum wells, and can reduce the density of InGaN MQW trench defects and the density of localization states induced by sever phase separation.[8,10] Phase separation causes uneven distribution of indium atoms, which affects the luminescent properties of LEDs.[11,12] However, the growth conditions of the barriers are rarely studied for yellow LEDs. Since the In composition is much higher than that of blue and green LEDs, growth conditions of the barriers impact significantly on the optoelectric properties of yellow LEDs.

In this Letter, to obtain high luminous efficiency yellow LEDs, we investigate the influence of growth temperature of barriers on the structure and optoelectric performance of yellow LEDs by electroluminescence (EL) spectroscopy, secondary ion mass spectroscopy (SIMS) and fluorescence (FL) microscopy, etc.

The InGaN/GaN yellow LED structure grown was obtained on $1.2 \times 1.2\,\mathrm{mm}^2$ patterned 2-inch silicon (111) substrates by a home-made metal organic chemical vapor deposition (MOCVD) reactor.[13] Figure 1 shows the epitaxial structure composed of 120-nm high temperature (HT) AlN buffer, then a 3-μm n-GaN layer, 6 nm LT-GaN, 32 periods of InGaN/GaN (5 nm/2 nm) superlattice (SL) layers, 10 nm LT-GaN, 9 pairs of yellow InGaN/GaN (3 nm/13 nm) MQWs (YMQWs), 50 nm Mg-doped $Al_{0.2}Ga_{0.8}N$, 230 nm p-type GaN.

Details of the growth process can be found elsewhere.[14] To facilitate the comparison, the same structure was designed for the four samples except the temperature of quantum barriers. The temperature of GaN quantum barriers of the four samples was set to

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be 910℃, 925℃, 930℃ and 940℃, which are for sample A, B, C and D, respectively.

After the epitaxial growth, using the reported chip manufacturing process,[15] the four samples are fabricated into the vertical structure LED chips in size of $1 \times 1\,\mathrm{mm}^2$ with the n-type GaN upwards and roughed. The EL test is characterized using the system consisting of IP250 integrating sphere and the CAS140CT spectrometer made by Instrument System. The temperature-controlling unit made by MMR Technologies, Inc. Chips with 562 nm is employed to control the wavelength under the forward current 350 mA in this work.

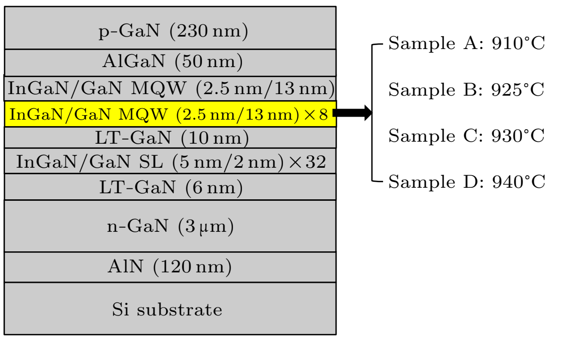

Fig. 1. Schematic of epitaxial structure of the samples with GaN barrier layers grown at different temperatures.

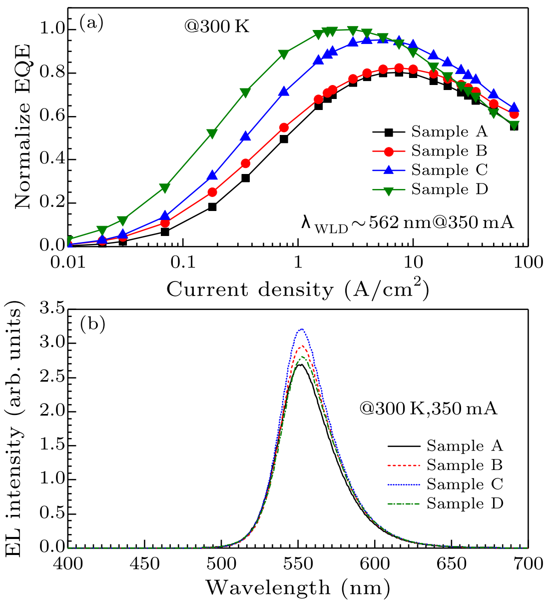

Fig. 2. (a) Normalized EQE versus current density of all the samples at room temperature, (b) EL peaks of the four samples at 350 mA and 300 K.

Figures 2(a) and 2(b) present the curves of normalized EQE as a function of the current density plotted in logarithmic scale under pulsed current injection to avoid heat efficiency at room temperature and the electroluminescence EL peaks of four samples at 350 mA and 300 K, respectively. It is observed that EQE increases when the barrier temperature increases from 910℃ to 940℃ at small current density. However, at large current density, the EQE increases with barrier temperature from 910℃ to 930℃, and then decreases when it varies from 930℃ to 940℃. Additionally, sample C owns the highest intensity of the EL peak in four samples.

Indium-element analysis of four samples was conducted by using a CAMECA IMS 7f secondary ion mass spectrometer (SIMS). As shown in Fig. 3, the larger the content of indium element, the steeper the barrier/well interfaces.[5] The detection depth between 250 and 400 nm corresponds to the quantum well region of the LED epitaxy structure. It can be seen from Fig. 3 that the content of indium in the quantum well region for sample C is largest among the four samples, indicating better barrier/well interfaces of sample C is expected. The interface quality of well/barrier of sample D is worse than that of sample C. One of the reasons is that too high barrier temperature is applied, and the indium atoms in the wells diffuse into the barriers,[16] leading to the deterioration of the well/barrier interfaces. The variation of the interface quality between the well and barrier may lead to the decrease of the external quantum efficiency of sample D.

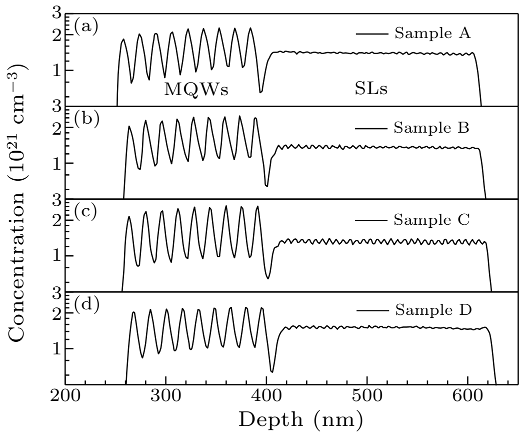

Fig. 3. SIMS compositional profiles of indium.

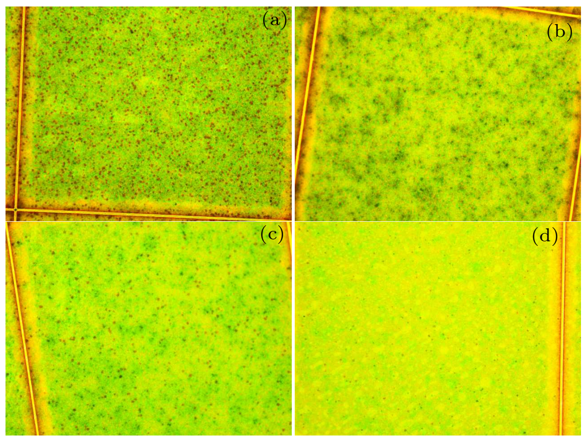

Fig. 4. Fluorescence microscopy images of epitaxial wafers of (a) sample A, (b) sample B, (c) sample C, and (d) sample D under the excitation of 420–490 nm intensive light.

The fluorescent luminescence (FL) images indirectly reflect the homogeneity of In composition in the InGaN quantum wells. Figure 4 shows the images of FL spectrometry for YMQWs excited by 420–490 nm excitation light source at room temperature.

Some “dark spots" from the FL image of four samples were observed by cathodoluminecence (CL) and microscopic photoluminescence (μ-PL), which may be caused by In clusters in the quantum well.[17–19] Hence, the number of the dark spots reflects the degree of phase separation in quantum wells. As shown in Fig. 4, with the increase of barrier temperature, the dark spots of the samples decrease, which indicates that the degree of In segregation in quantum wells decreases. Thus, increase in quantum barrier temperature can suppress effectively the generation of phase separation.

The generation of phase separation is the result of very low mutual solubility and large lattice mismatch between InN and GaN, which causes the transverse inhomogeneity of In component in quantum wells and the formation of In-rich local state (quasi-quantum dots).[20–22] This localized state plays a three-dimensional limiting role on carriers, making it more difficult for carriers to migrate to non-radiative recombination centers caused by defects, thus greatly improving the efficiency of radiation recombination luminescence. Therefore the decrease in the EQE of sample D may be related to the weakening of the local state.

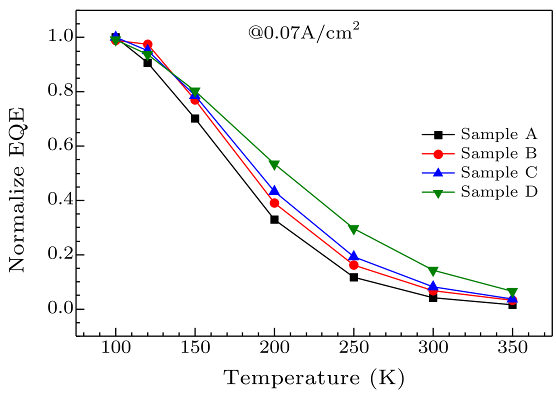

Fig. 5. When the injection current density is 0.07 A/cm^2, the EQE curves of samples A, B, C and D vary with temperature. In order to facilitate comparison, EQEs of the four samples have been normalized at the same current density.

Figure 5 gives the EQE curves for sample A, B, C and D with various temperatures (100–350 K) at the low injection current density of 0.07 A/cm^2. For comparative purposes, the EQE data of the four samples at 100 K were normalized at the same current density. It can be seen that the EQE of sample A decreases more severely than those of the other three samples with the increase of temperature, which is the so-called temperature droop (T-droop). The T-droop of the GaN-based LED under low current density injection is caused by defect-related non-radiation (SRH).[23–25] The defects in the active region of QWs are more sensitive to small current density than to large current density. When the temperature increases, the defects will be thermally activated. The lifetime of carriers will be reduced.[26] The more the defects in the active region of QWs, and the more serious the T-droop under the condition of small current density injection. Thereby, the degree of temperature droop under low current density injection can reflect indirectly the crystal quality in the QWs.

The $J_{\rm max}$ is defined by the current density corresponding to the peak value of EQE shifts toward higher current density as the barriers temperature increases (see Fig. 2(a)), and the lower $J_{\rm max}$ can be interpreted as longer non-radiative lifetime and better crystal quality of the InGaN quantum well. Therefore, raising barrier temperature will decrease the defects in the active region of QWs, and the crystal quality is improved. Obviously, sample D possesses the highest EQE at low current because of the good crystal quality of InGaN QWs.

Four GaN-based yellow LEDs with different growth temperatures of quantum barrier grown on Si substrates have been designed and manufactured in this work. The effect of the growth temperature on the structure and photoelectric performance of the yellow LED is investigated by EL spectroscopy, SIMS and FL microscopy. It is found that with the increase of barrier temperature, the crystal quality of QWs and the interface quality of the well/barrier are improved effectively, and the quantum well is thermally annealed, so that the indium atoms in the quantum well migrate to the equilibrium position, reducing the phase separation of the quantum well and improving the crystal quality of the quantum well. Therefore, the photoelectric performance of the yellow LED is greatly improved. However, the barrier temperature continues to increase, and EQE and EL intensity begin to decrease. There may be related to the weakening of the local state and the deterioration of quality of the well/barrier interface.

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