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Effect of annealing on photoluminescence and microstructures of InGaN/GaN multi-quantum well with Mg-doped p-type GaN*

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InGaN/GaN multi-quantum well structure with Mg-doped p-type GaN was grown by low-pressure metalorganic vapour phase epitaxy. After rapid-thermal-annealing at 700 and 900°C, both the red-shift and the blue-shift of the photoluminescence (PL) peak, the decreased and the enhancement of the PL intensity were observed. The transmission electron microscopic images showed that InGaN multi-quantum-dots-like (MQD-like) structures with dimensions less than 5×10nm were formed in InGaN wells. The changes of PL spectra could be tentatively attributed to the competition between the red-shift mechanism of the quantum-confined Stark effect and the blue-shift mechanism of the quantum size effect due to MQD-like structures.

Keywords: InGaN, rapid-thermal-annealing, multi-quantum well
PACC: 7865K, 8140T

1. Introduction

The Mg-doped p-type GaN is widely used in preparation of optoelectronic devices, such as blue, green and ultraviolet light emitting diodes (LEDs) and laser diodes. But the as-grown Mg-doped GaN generally shows a high resistivity due to the large hydrogen passivation of the Mg acceptors.[1] The p-type conductivity of Mg-doped GaN is mainly dependent on the growth and annealing (or activation) conditions.[2] In general, the major parameter of the forward voltage of GaN-based LEDs is closely related to the conductivity of the p-GaN attained at different annealing temperatures.[3] At the same time, for the InGaN/GaN multi-quantum well (MQW) structure, the interfaces between InGaN wells and GaN barriers are also affected due to the interdiffusion of In and Ga atoms. Therefore, the annealing is a crucial process for the optical properties and interfacial microstructures of the InGaN/GaN MQW structure with Mg-doped p-type GaN.[4]

In this work, the InGaN/GaN MQW structure with Mg-doped p-type GaN is grown by means of low-pressure metalorganic vapour phase epitaxy (LP-MOVPE). The influence of rapid-thermal-annealing (RTA) on optical properties and distributions of constituents in this structure is investigated using photoluminescence (PL) and transmission electron microscope (TEM). Especially, the effect of the multiple-quantum-dot-like (MQD-like) structure in InGaN wells on the peak wavelength and intensity of the PL spectra is studied in detail.

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2. Experiments

The InGaN/GaN MQW structure with Mg-doped p-GaN was grown by LP-MOVPE on a (0001) sapphire substrate. High-purity H_2 and N_2 were used as carrier gases. Trimethylgallium (TMGa), trimethylindium (TMIn), trimethylaluminium (TMAl), and ammonia (NH_3) were used for gallium, indium, aluminium, and nitrogen precursors, respectively. Saline (SiH_4, 100ppm in H_2) and biscyclopentadienyl-magnesium (Cp_2Mg) were employed as the n-type and p-type dopants, respectively. The procedure of material growth was as follows. At first, the sapphire was treated at 1150°C in H_2 for 10min. Then, a 20nm GaN nucleation layer was deposited on the sapphire at 550°C. After this, a 1μm unintentionally doped GaN and 2μm n-type GaN were grown at 1040°C. The five-period MQW structure, consisting of 2nm InGaN wells and 8nm GaN barriers, was then grown on the n-GaN layer at different growth temperatures around 800°C. The low and high temperatures were helpful to grow the InGaN wells and the GaN barriers in MQW, respectively. Therefore, a suitable growth temperature should be selected for high-quality MQW structure growth. Finally, a 0.2μm p-type GaN was grown at 1060°C.

The samples, annealed at 700°C and 900°C in N_2 for 20 min, were denoted as sample A and sample B, respectively. The PL spectra were measured at room temperature, and the excitation source was a He-Cd laser with 325nm line. The TEM samples were prepared by using the standard procedure including Ar^+ ion thinning.

3. Results and discussion

The PL spectra of the as-grown samples, sample A and sample B, are shown in Fig.1(a), and the effect of annealing temperature on the PL peak intensity is shown in Fig.1(b). The dominant peak of the as-grown samples is located at about 440nm at room temperature. For sample A, the PL peak wavelength (453nm) is red-shifted by 13nm and the intensity decreases, as compared with the as-grown sample. In contrary to sample A, the PL peak wavelength (449nm) of sample B is blue-shifted by 4nm and the intensity increases obviously. Especially, for sample B, the PL peak wavelength is shorter and the intensity is much higher than that of the as-grown one.

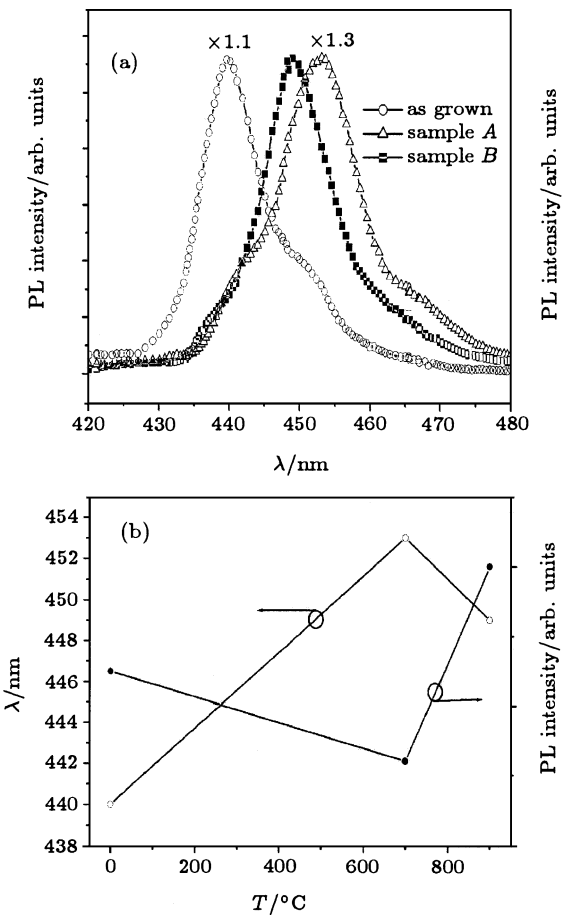


Fig.1. Effect of annealing on PL spectra of as-grown sample, sample A and sample B.

In principle, the quantum-confined Stark effect (QCSE) would be reduced in InGaN/GaN MQW structure after annealing, resulting in a blue-shift of the PL peak wavelength. But our experimental results are not consistent with the above-mentioned blue-shift caused by annealing.

For the as-grown sample, the TEM image showing the cross-section of the interfacial microstructures between InGaN wells and GaN barriers is shown in Fig.2. The distinct and regular interfaces between InGaN wells and GaN barriers are identified in the MQW structure.

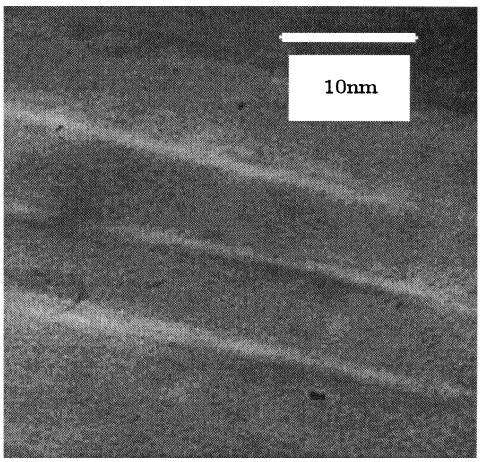


Fig.2. The TEM image showing the cross-section of the as-grown sample.

In the TEM image, as shown in Fig.2, fewer dark-dot structures could be seen in the InGaN wells for the as-grown sample. In addition, for sample *A* (700°C/20min), the thickness of InGaN wells was broader than that of as-grown sample.[5] The thickness change of InGaN wells was caused by the interdiffusion of In and Ga atoms between InGaN wells and GaN barriers.[4] Therefore, the PL peak wavelength became red-shifted with the decrease of the quantum confinement effect due to the broadening of InGaN wells after annealing.

The TEM image showing the cross-section of sample *B* (900°C/20min) is shown in Fig.3. The image reveals that the interfaces between InGaN wells and GaN barriers are very rough and disordered. Many dark-dot structures can be clearly seen in the InGaN wells.

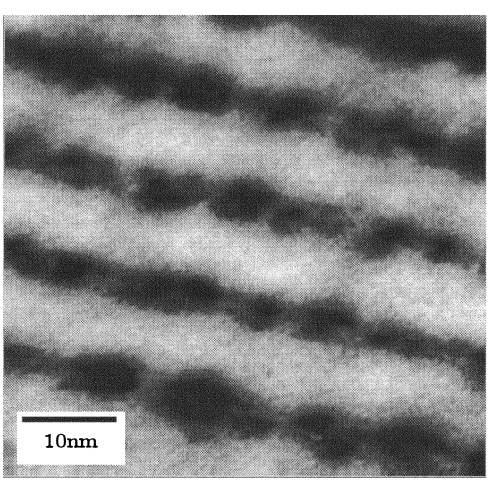


Fig.3. The TEM image showing the cross-section of sample *B* (900°C/20min).

The In composition in the dark-dots is higher than that in other regions as disclosed by the energy dispersive x-ray analysis, therefore, the dark-dot and relatively lighter regions in InGaN wells can be attributed to In-rich and In-poor phases,[6,7] respectively. These dark-dots originate from the formation of the In-rich phase, which is caused by the fluctuation of In content in the InGaN wells. The above results indicate that annealing at 900°C could facilitate the formation of the dark-dot In-rich phase in the InGaN wells.

The dark-dot structure of In-rich phase is so-called quantum-dot-like (QD-like) structure, and really exists in the InGaN wells. So, it is named MQD-like structure because of the large number of dark-dots.

As shown in Fig.3, the distribution of In content in the wells tends to be concentrated. The interfaces between wells and barriers are quite irregular although their boundaries are clearly observed. Such a gathering process of In concentration, caused by spinodal decomposition,[8] induces a quasi-regular QD-like structure with In-rich phase in the previous wells.[9] Here, we could think that the InGaN wells become MQD-like layers, namely, the MQW structure changes into a MQD-like structure through annealing at high temperatures. The sizes of MQD-like layers are less than 5nm×10nm. So, its thickness was much greater than that of the InGaN wells annealed at 700°C, possibly due to the interdiffusion of In and Ga atoms between InGaN wells and GaN barriers.

Generally, the QD-like structures with In-rich phase can be regarded as deep traps, which can capture electrons and holes. Then the carriers are deeply localized in the QD-like structures in three-dimensional (3D) confinement. Thus, a strong band-to-band transition becomes the dominant emission mechanism, and the peak wavelength and the intensity of the PL spectra of the InGaN/GaN MQW structure can obviously be improved.[10,11]

In this work, the variation of the PL spectra could be tentatively attributed to a competition between the red-shift mechanism of QCSE and the blue-shift mechanism of the quantum size effect including MQD-like structures. As mentioned above, we proposed that the quantum size effect of MQD-like structures becomes the dominant emission mechanism in InGaN/GaN MQW blue LEDs annealed at high temperature.

4. Conclusion

In summary, for the InGaN/GaN MQW blue LED annealed at high temperatures, it was found from TEM micrographs that the MQW could be changed to MQD-like structure. The influence of MQD-like structure on the emission mechanism could give rise to a change in the optical properties of blue LEDs such as wavelength and intensity of the PL spectra. Thus, the quantum size effect, including MQD-like structures, becomes the dominant emission mechanism in InGaN/GaN MQW blue LEDs annealed at high temperatures.

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