Influence of Rapid Thermal Annealing on the Characteristics of InGaN/GaN MQWs

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Abstract. N-type InGaN/GaN multiple-quantum-wells (MQWs) were grown on sapphire substrates by metal organic chemical vapor deposition (MOCVD). The crystal quality and optical properties of samples after rapid thermal annealing (RTA) at different temperatures in a range from 400 to 800℃ are investigated by X-ray diffraction (XRD) and photoluminescence (PL) spectrum. The experimental results show that the peaks of InGaN, InN and In can be observed in all samples. And the results are induced by the phase separation and In-clusters. The luminescence peak of the samples annealed showed a red shift. It is caused by strain stress relaxation during the RTA process. Furthermore, some defects can be eliminated and the best annealing temperature is from 500℃ to 700℃.

Keywords: InGaN/GaN MQWs, RTA, XRD, PL spectrum

1 Introduction

Due to its high luminescent efficiency, high stability and changeable emission wavelength, InGaN/GaN MQWs, working in broad spectrum (from ultraviolet to infrared) as active layers of light emitting diode (LED) luminescence devices, is an excellent semiconductor luminescence material[1,2,3]. This kind of MQWs semiconductor is of great interest for the researchers in recent years. At present, the growth temperature dependence of InGaN/GaN MQWs grown by MOCVD for photoluminescence was experimentally investigated [4]. However, the relative reports on thermal annealing were few, especially RTA. Because of the distribution of In-composition and the strain stress relaxation between well layer and barrier layer in InGaN/GaN MQWs, the optical properties can be changed after these thermal treatments. The strain stress relaxation can decrease piezoelectric field, and the other properties of MQWs will be different with the variation of piezoelectric field. In this paper,

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the behavior of InGaN/GaN MQWs at different RTA temperatures was studied by means of PL and XRD.

2 Experimental

The InGaN/GaN MQW devices were grown by MOCVD on c-plane sapphire substrate. It was composed of a 30nm thick n-type GaN layer grown at 1050℃, a ten-period InGaN(6nm)/GaN(8nm) MQW grown at 750℃, and a 200nm p-type GaN layer grown at 960℃. During the growth, Trimethylgallium (TMGa), Trimethylindium (TMIn) and ammonia (NH_3) were used as Ga, In, and N precursors, respectively.

The samples size is $5\times5\ mm^2$. One-step RTA processes were performed on the samples with a heating rate of 80℃/s at 400,500,600,700 and 800℃for 180s respectively in N_2 atmosphere. X-ray rocking curves with InGaN (0002) diffraction were obtained by CuKα1 irradiation. The optical properties of samples were investigated at room temperature by PL using a 325nm He-Cd laser as the excitation source.

3 Results and Discussion

Fig.1 shows the X-ray rocking curve at room temperature of as-grown InGaN sample. The highest peak at 34.66° is GaN diffraction peak, InGaN(0)–certificate diffraction peak is at 34.57°. And the diffraction peak at 34.45° is InGaN(-1)–certificate peak. There is a broad distance between the two diffraction peaks of InGaN. It is caused by the In-composition fluctuation in different regions. There are two lower peaks on the leftmost in Fig.1, one is diffraction peak of InN at 32.71° and the other is In diffraction peak at 33.52°.

The In diffraction peak is ascribed to segregation phenomenon of indium on the surface during the growth. First, the absorbability of indium atom on substrate is increased. Second, decomposition efficiency of NH_3 get faster, the concentration of nitride is increased. These factors are beneficial to In-N bond formation. However, the appearance of InN diffraction peak is owing to phase separation which caused by indium segregation phenomenon.

That is to say, In-composition fluctuation and the order distribution of indium atom may cause the appearance of InN. And In-cluster produced by In-composition fluctuation and phase separation can emerge small potential well. It can make carriers bound and reduce the capture probability of unirradiative recombination centre, luminescence efficiency has been enhanced. In general, even though InGaN/GaN MQWs has bigger dislocation density, it has higher luminescence quantum efficiency [5]. Therefore, In-composition fluctuation and phase separation are significant.

On the structure of InGaN/GaN MQWs, the Bragg angle of InGaN(0)–certificate diffraction peak is the average Bragg angle in the whole quantum wells[6]. According to the Bragg formula and hexagonal crystal inverted space formula, it can be calculated the horizontal (a=0.3225nm) and perpendicular (c=0.5267nm) average crystal lattice constants according to Eq. 1 and Eq. 2 in InGaN/GaN MQWs, respectively.

$$2d\sin\theta_B=\lambda \tag{1}$$

$$d_{hkl}=\frac{1}{\sqrt{\frac{4}{3}\left|\frac{h^2+hk+k^2}{a^2}\right|+\left|\frac{l^2}{c^2}\right|}} \tag{2}$$

Where $\theta_B$is the Bragg angle of InGaN(0)–certificate diffraction peak, $\lambda=0.514nm$ is the wavelength of X-ray [7]. For the epitaxial sample of InGaN/GaN MQW, it is in the

undelivered strain state, and the In-composition (x=0.3) can be calculated by the following formula [8],

$$x=-\frac{ac\left(1+\nu\right)-ac_0^{GaN}-a_0^{GaN}c\,\nu}{ac_0^{GaN}-ac_0^{InN}-a_0^{InN}c\,\nu+a_0^{GaN}c\,\nu} \quad (3)$$

Where ν=0.6 [9] is Poisson coefficient which is in hexagonal crystal. a_0^{GaN}=0.3189nm and c_0^{GaN}=0.5185nm are the horizontal and perpendicular theoretical crystal lattice constants in GaN, respectively. a_0^{InN}=0.3533nm and c_0^{InN}=0.5693nm are the horizontal and perpendicular theoretical crystal lattice constants in InN, respectively. The calculation result of In-composition (x=0.27) is very closely with the growing sample.

Fig.2 shows the PL spectrum at room temperature of as-grown InGaN. The wavelength of emission peak of InGaN is at 441nm with full width at half maximum (FWHM) value of 46nm. In addition, the peak of GaN with wavelength of 388nm is also observed. The peak of yellow band (YB) has not been obtained from Fig.2, which indicates the crystal perfection of sample is higher.

As shown in Fig.3, there are six obviously shoulder peaks of the samples annealed. However, because of a large lattice mismatch between GaN and InGaN, the relaxation phenomenon will occur immediately when InGaN grown on GaN. These peaks may well correspond to the defects. Then the dislocation density in sample is increased with the decreasing of crystal quality. With the increasing of annealing temperatures, the luminescence intensity of emission peak gets stronger. It indicates that some defects can be eliminated after annealing [10].

Moreover, Fig.3 indicates that the luminescence peaks of InGaN/GaN MQWs show a red shift imperceptibly after RTA, which annealing at different temperatures for 180s. The results can be explained by the strain stress relaxation during the RTA, which can consistent with the results of X-ray rocking curves. Samples annealed at high temperature, gallium and indium atom diffusion rate increases, crystal lattice mismatch release internal strain stress. So the piezoelectric field-effect of MQWs decreases, and it also has influence on luminescence properties. At the same time, intrinsic defects in MQWs and phase separation caused by In-composition diffusion-limited showed a red shift.

Fig.4 shows the intensity of emission peaks of samples annealed at different temperatures. There are almost no differences from 500℃ to 700℃. But the intensity has been increased from 700℃ to 800℃ suddenly. As the same time, the emission peak of sample annealed at 400℃ is very closely to as-grown sample. Fig.5 shows the intensity of shoulder peaks of samples about 706nm annealed at different temperatures. With the increasing of RTA temperatures, the intensity of shoulder peaks has been decreased. Furthermore, it can be seen that the expressions range from 400℃ to 700℃ are the lowest, and then it has been greatly increased at 800℃. Therefore, we can make a conclusion that there are some new defects of sample annealed at 800℃. So the best annealing temperature is from 500℃ to 700℃ for defects elimination. In-composition and InN phase segregation has been changed after RTA, which influences the PL spectrum of samples.

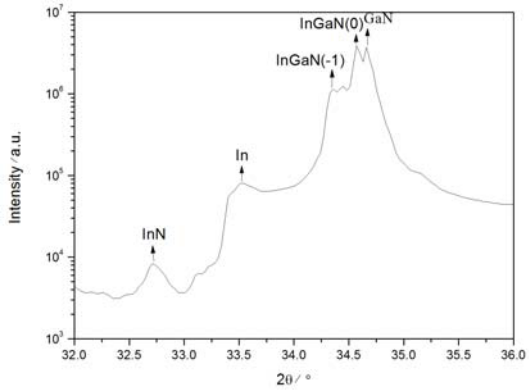

Fig.1 X-ray rocking curve of as-grown InGaN sample

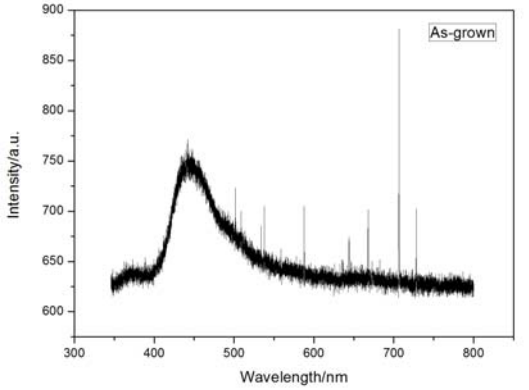

Fig.2 the PL spectra of as-grown InGaN measured at room temperature

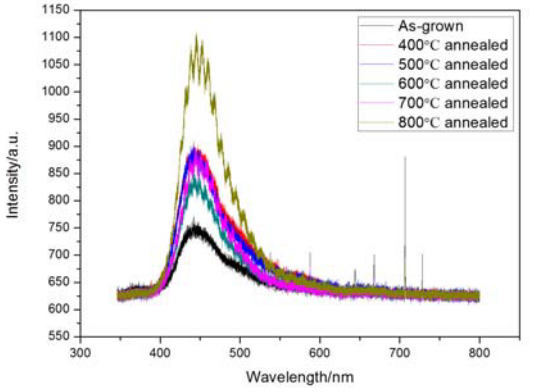

Fig.3 PL spectra of InGaN annealed at different temperatures

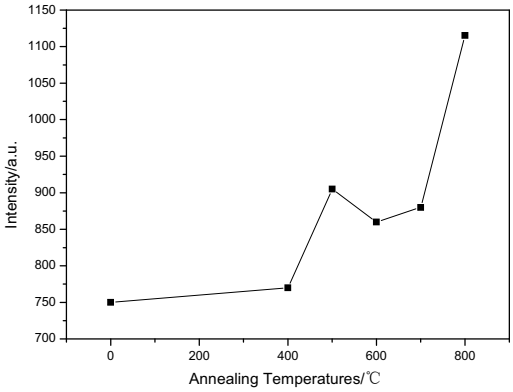

Fig.4 Intensity of emission peaks of samples annealed at different temperatures

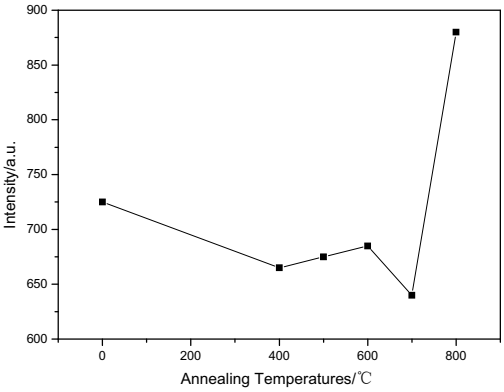

Fig.5 Intensity of shoulder peaks at 706nm of samples annealed at different temperatures

4 Conclusions

According to the results of the X-ray rocking curve at room temperature of as-grown InGaN sample, the average In-composition can be calculated accurately, which is almost consistent with actual growth conditions of InGaN/GaN MQWs. And the peaks of InGaN, InN and In are observed on XRD spectrum. It is the evidence of phase separation and In-clusters in the sample. Furthermore, RTA process can eliminate some defects properly and the best annealing temperature is from 500℃ to 700℃. It indicates that the energy band of MQWs can be changed by annealing. At the same time, with the variation of In-composition and phase separation, the luminescence properties are changed.

Acknowledgements

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