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The Structure and Optical Performance of InGaN/GaN Multiple Quantum Wells Grown on C-plane Sapphire

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Abstract

In this article, we study the structure and optical performance of penetration dislocation to the epitaxial GaN-base quantum well on C-plane sapphire. On the one hand, the penetration dislocation will make the interface of quantum wells disperse and largely fluctuate, and accordingly increase the localization effect of quantum wells. On the other hand, as the nonradiative recombination center, the penetration dislocation makes the radiation intensity of quantum wells to be reduced. The influence of penetration dislocation to the optical performance of InGaN/GaN multiple quantum wells decides the competition between both. In addition, the localization effect intensity of InGaN quantum wells possesses linear relation with the content of In in definite range of wavelength.

Keywords: Multiple quantum wells, Metal organic chemical vapor deposition, X ray, AFM

1. Introduction

In this article, we widely and deeply study the growth condition, structure character and radiation performance of InGaN/GaN structure. But because the crystal lattice mismatch of InN and GaN is big, the solid solubility is small and the epitaxial growth condition is very rigorous, so the quality of crystal is low. In the process of growth, the alloy of InGaN will produce the phenomenon of mutual separation, form the enrichment zone of In similar with quantum point, and bring the localization effect to the carrier (Yukio Narukawa, 1997, P. 1938 & P. 981). And the dislocation density on the epitaxial layer is high (10^8-10^{12}/cm^2), and the GaN-base LED grows along the direction of [0001], and the spontaneous polarization effect and the piezoelectricity polarization effect exist in the nitride from this direction, and these factors are integrated together, which makes the problem more complex. So for the InGaN/GaN quantum wells, there are many problems to be further studied.

Differing with the epitaxies of other compound semiconductor system, Ⅲ-V nitride can only be extended on other underlay with big mismatch and heterogeneity. The crystal lattice mismatch of GaN and sapphire is 14%, and the big mismatch inevitably induces two basic problems in the epitaxial films, the deficiency with high density and the large plane stress. We can adopt the two-step growth method, and the penetration dislocation density is 10^8-10^{12}/cm^2, but surprisingly, though such high dislocation density exists, the exterior quantum efficiency of GaN-base LED has achieved 30% at present, and the life has achieved tens thousands hours, which is unimaginable in arsenide semiconductor and phosphide semiconductor and arouses human large interests, and people have deeply researched the dislocation character in the GaN epitaxial material.

2. Experiment

In the experiment, we select the common C-plane sapphire (0001) as the underlay, utilize the low-pressure MOCVD system to grow the LED structure with multiple quantum wells, and get the epitaxial films with different penetration dislocation densities through the change of growth condition. And we utilize temperature-change fluorescence, AFM, high resolution X ray diffraction and other measures to study the influences of penetration dislocation to the structure and optical performance of InGaN/GaN multiple quantum wells.

Before the epitaxial material grows, in hydrogen, the sapphire underlay is heated to 1050℃ in ten minutes to eliminate the impurities on the surface of the underlay. The epitaxial growth adopts the two-step growth method, and first grow the GaN low temperature buffer layer with the depth of 30nm in 500-600℃, through 1000℃ high temperature anneal, grow the LED structure under high temperature. The structure includes 4μm GaN:Si under 1050℃, InGaN/GaN multiple quantum wells with 5 periods grown under 800℃, and the p area grown on 1000℃. The GaN barrier of quantum wells is mixed into Si, and the adulteration concentration is 3×10^{18} cm^{-3}, and InGaN is not adulterated. The structures of sample A and sample B are same in the experiment, and the difference is that both samples adopt different Ⅴ/Ⅲ ratio (the Ⅴ/Ⅲ ratio of sample A is 1280 and the Ⅴ/Ⅲ ratio of sample B is 640) to grow ten minutes in the initial growth stage of high temperature GaN, then both samples adopt the Ⅴ/Ⅲ ratio of 1280 to complete the growth of high temperature GaN layer. In the growth process, we adopt the method of optical measure to implement original position supervision, and the detection light wavelength is 600nm to optimize the growth conditions.

In the measure of fluorescence, the blaze lamp-house adopts 7mW He-Cd laser with 325nm, the system adopts 0.5m homochromy meter and GaAs photoelectrical multiple increase detector, and the temperature range of temperature change measurement is 10K-300K. The spectrum curve eliminates the influence of F-B interference through Lorenz fitting, and we can get exact fluorescence peak value and integral intensity. High resolution X ray diffraction is measured by the Bede D1 system, and the wavelength of X ray is λ=0.1541nm. After measuring the fluorescence character of epitaxial structure, we utilize the method of plasma etch to eliminate the p area and multiple quantum wells area, and then put the samples in the phosphoric acid of 160℃ to erode in 6 minutes, then study the dislocation density and character of epitaxial films, and the disposal detail is in P. Visconti's article (P. Visconti, 2002, P.229).

3. Result and discussion

The dislocation density of the etched sample is confirmed by AFM measurement. The measure range of AFM is 3μm×3μm, and the image is seen in Figure 1. The maculas in the image are sunken holes eroded by the phosphors acid when the penetration dislocation is terminated on the surface of films (D. Kapolnek, 1995, P.1541). Through the measure of the density of maculas, we can calculate the penetration dislocation density of epitaxial material. From the Figure, we can see that the dislocation densities of etched sample A and sample B (they are signed as BLA and BLB respectively) are respectively $1.6\times10^{9}cm^{-2}$and $7\times10^{7}cm^{-2}$, and comparing with BLA, the dislocation density of BLB is lower.

Because the dislocation density of BLA is high, so the etched surface is coarser than BLB. In the GaN epitaxial process of two-step growth method, though the quality of low temperature buffer layer crystal is bad, but it reduces the interface energy between GaN and sapphire, and offers an easy core-formation surface for the growth of high temperature GaN. The high temperature GaN first forms the GaN island on the buffer layer, and in the initial growth term, the GaN island continually grows up till encounter, and forms continual epitaxial films, and the growth is translated into the step-flow mode, and the GaN films are transformed into smooth films which surfaces have many steps. Strictly speaking, the epitaxial films of GaN is a sort of structure similar with mosaic, here, the penetration dislocation is composed by two parts, and the small part is in the middle of the island, and the big part is at the boundary encountered by islands. So the growth condition of high temperature GaN in the initial growth stage has decisive influences to the crystal quality of the whole epitaxial films. In this stage, most part dresses are released, and with the encounter and mature of islands, the dislocation produces at the boundary of islands. And the researches indicate the Ⅴ/Ⅲ ratio can effectively influence the size of island in the initial growth period of epitaxial material, and change the dislocation density in the materials (T. Yang, 2000, P.45 & S. Figge, 2000, P.262), and the experiment result approves that. Because the Ⅴ/Ⅲ ratio in the initial growth period of high temperature GaN is reasonable selected, the dislocation density of sample B reduces over one quantity level.

The swing curve (ω scan) of X ray diffraction includes symmetry scan and asymmetry scan, and it is a sort of method which is extensively used to study the deficiency of epitaxial films, and the peak width at half height of swing curve can reflect the density and character of dislocation. Figure 2 displays the X ray ω scan symmetry (002) and asymmetry (105) curves of BLA and BLB, and the fitting results of experiment data show that the half peak widths of symmetry (002) and asymmetry (105) scan of BLA are respectively 331.8arcsed and 356.3aresec, and corresponding fitting data of BLB respectively are 244.8arcsed and 252.1aresec. We can see that the swing curve of asymmetry scan is more obvious than the symmetry scan. According to B. Heying's researches (B. Heying, 1996, P.643), for the symmetry scan mode, only the whorl dislocation and mixed dislocation in the epitaxial layer would produce the expanding width with peak shape, and the pure edge dislocation has no influence to the expanding width with the peak shape, and for the asymmetry scan mode, all types of dislocation will produce the expanding width with peak shape. In addition, according to the researches of the fitting to the GaN (002) diffraction peak, we found its linearity was close to pure Gauss linearity, which indicates that the epitaxial films possess the structure of mosaic, and the expanding width of diffraction peak is mainly aroused by mutual function of crystal epitaxial warp, transverse size effect, and epitaxial films location press field. From the half peak widths of two samples, the half peak width of BLB is small, and it presents better crystal quality, which accords with the result of AFM measure.

In conclusion, the measures of AFM and X ray swing curve all prove the optimization of the Ⅴ/Ⅲ ratio in the initial high temperature GaN growth period can effectively reduce the dislocation density of epitaxial material and enhance the crystal quality and optical performance of material. Based on GaN epitaxial materials with different dislocation densities, we studied the influences of penetration dislocation to the structure and optical performance of InGaN quantum wells.

Generally speaking, the measure of X ray macle diffraction to the epitaxial single crystal film usually adopts two sorts of scan mode, i.e. the θ-2θ scan mode that the sample circumrotates θ angle and the detector circumrotates 2θ angle, and the ω scan mode that the detector is fixed on the position of double diffraction angle and the sample only circumrotates about the diffraction angle, i.e. the swing curve. Simply speaking, the θ-2θ scan is sensitive to the crystal lattice aberration of crystal at the vertical direction on the surface, and the ω scan is sensitive to the aberration of crystal at the parallel direction on the surface.

To further ravel the influence of penetration dislocation to the optical performance of multiple quantum wells, we utilize the temperature-change fluorescence to study two samples, and the temperature range of measure is 10k-300k.

Some researches indicate that GaN-base LED possess thus high radiation efficiency because the phenomenon of mutual separation will occur in InGaN alloy and the enrichment zone of In similar to quantum points forms, which will produce the localization effect to carriers and be propitious to the compound radiation of electron and cavity. So the radiation of InGaN quantum wells shows the localization characteristics. Because large numbers of enrichment In areas exists in quantum wells and the components of In in different enrichment In areas are different, so to electron and cavity, different potential energy minimums exist, which is seen in Figure 3. Under very low temperature, the heat excitation energy is small, and the radiation carrier can only be randomly distributed at different potential energy minimum values and form localization exciton, and the nonradiative compound and heat effect can be ignored.

Figure 3 shows fluorescence spectrums of sample A and B quantum wells under 10k of low temperature in the radiation area, and the radiation peak values displayed on two spectrum lines are about 2.68eV (463nm). On the spectral side with low energy, the extension of sample A is little higher than the extension of sample B, and the linearity forms of two spectrum lines are consistent. But on the side with high energy, the extension of sample A is much higher than sample B. InGaN quantum wells under low temperature is localization exciton compound radiation, the high energy end and the low energy end of spectrum line respectively reflect the compounds of radiated carriers on the weak localization energy level and on the strong localization energy, which indicates the penetration dislocation inducts shallow localization energy level, and influences little the deep localization energy level. The localization effect of InGaN quantum wells mainly roots in two aspects, i.e. the mutual separation of InGaN alloy and the width fluctuation of quantum wells formed by the interface fluctuation of quantum wells. According the analysis of HRXRD, the quantum wells interface fluctuation of sample A is bigger than sample B, so we can think that the shallow localization energy level roots from the fluctuation of quantum wells interface.

With the further increase of temperature, the heat energy inspires the carriers from the localization energy level, and more and more carriers are seized by the nonradiative compound center, and the nonradiative compound process begins to increase until it occupies the leading status. Because the penetration dislocation density of sample A is higher than sample B, and the attenuation of sample A is more notable than sample B in this temperature area, which can prove that the penetration dislocation certainly is effective nonradiative compound center in InGaN quantum wells.

The differences of light intensity and peak form with different radiation wavelengths for the quantum wells are produced by the differences of the crystal quality and In content of InGaN alloy, and the differences of growth temperature and In component for the quantum wells will produce the localization differences of quantum wells. Figure 4 shows the relationship between the radiation wavelength and In component of quantum wells and the localization energy, and with that the radiation wavelength of quantum wells increase from 365nm and 460nm to 516nm, the In content in the InGaN alloy presents ascending trend which increase from 3.1% and 9.6% to 17.6%, and the localization energy of quantum wells also present ascending trend, and both aspects possess linear relationship. It is obvious that the In component and localization energy of quantum wells have same change trend in this radiation wave band, which proves that the localization effect of quantum wells is mainly produced by the participation of In atoms.

From above experiment results, we can see that the influences of penetration dislocation to the structure of optical performance of quantum wells include flowing aspects, and the first one is that the penetration dislocation will deteriorate the interface quality of quantum wells and introduce the localization effect in InGaN quantum wells, and the second one is that the penetration dislocation is the nonradiative compound center in the quantum wells and makes the radiation intensity of InGaN quantum wells reduced.

4. Conclusions

In this article, we study the influences of penetration dislocation to the structure and optical performance of epitaxial GaN quantum wells on C-plane sapphire. And the research found that on the one hand, the penetration would disperse the interface of quantum wells, and increase the fluctuation, and accordingly increase the localization effect of quantum wells, and on the other hand, as the nonradiative compound center, the penetration dislocation would reduce the radiation intensity of quantum wells. The influences of penetration dislocation to the optical performance of InGaN/GaN multiple quantum wells are decided by both competition.

The localization effect intensity of InGaN quantum wells possesses linear relationship with In content in certain wavelength range. The In content has important influence to the light intensity with the attenuation of temperature, but high In content will weaken the crystal quality of quantum wells, and comparing with purple light and green light, the blue light radiation possesses better optical performance, which is the balanced result between two aspects.

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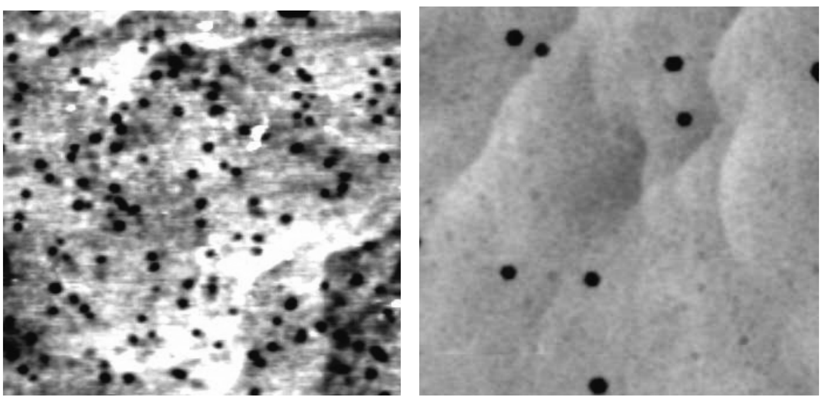

(a) (b)

Figure 1. AFM Images of Sample (a) and Sample (b) after Etching (The Measure Area is 3μm×3μm)

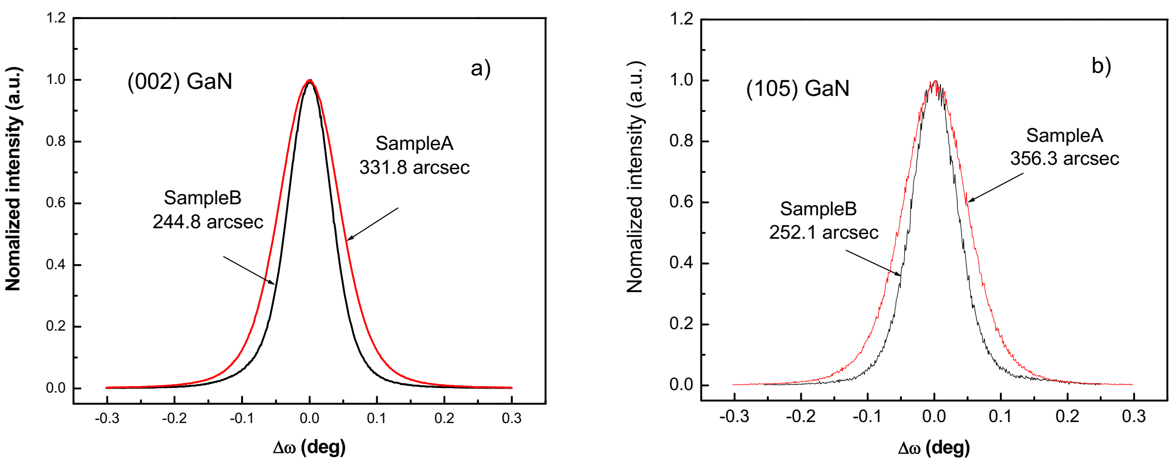

Figure 2. X-ray ω Scan Curves of Sample Symmetry (002) and Asymmetry (105)

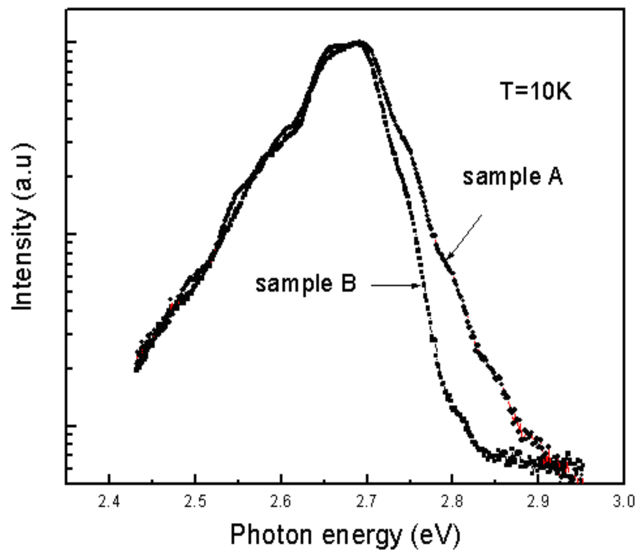

Figure 3. Quantum Wells Low Temperature PL Spectrum of Sample A and B

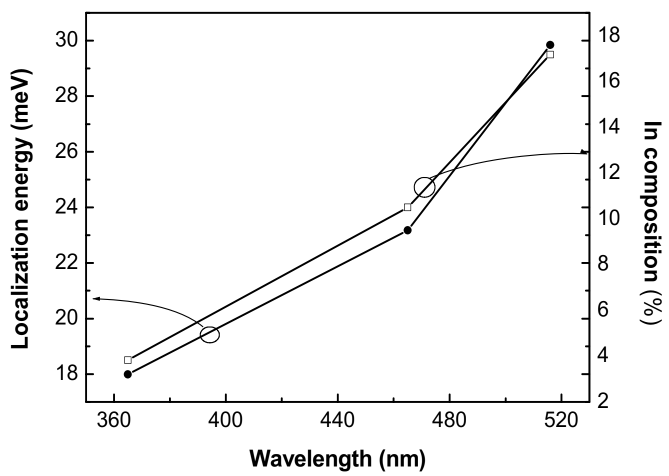

Figure 4. The Relationship of Quantum Wells Radiation Wavelength and In Component with Localization Energy