OPTICAL PROPERTIES OF Si-DOPED $Al_xGa_{1-x}N/Al_yGa_{1-y}N$ (x=0.24-0.53, y=0.11) MULTI-QUANTUM-WELL STRUCTURES

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ABSTRACT

We demonstrate strong ultraviolet (UV) (280-330nm) photoluminescence (PL) emission from multi-quantum-well (MQW) structures consisting of AlGaN active layers fabricated by metal-organic chemical-vapor-deposition (MOCVD). Si-doping is shown to be very effective in order to enhance the PL emission of AlGaN QWs. We found that the optimum values of well thickness and Si-doping concentration of $Al_xGa_{1-x}N/Al_yGa_{1-y}N$ (x=0.24-0.53, y=0.11) MQW structure for efficient emission were approximately 3nm and $2\times10^{19}cm^{-3}$, respectively. In addition, the PL intensities of AlGaN, GaN and InGaN quantum well structures are compared. We have found that the PL emission at 77K from a $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ MQW is as strong as that of InGaN QWs.

INTRODUCTION

GaN and related nitrides are currently of great interest for the application to optical devices operating in the visible and ultraviolet (UV) energy range. Blue laser diodes (LDs) and blue-green light-emitting diodes (LEDs) have been developed in recent years [1-3]. High-power, long-lifetime InGaN multi-quantum well (MQW) lasers have been demonstrated [1].

The AlGaN alloy is a useful material for optical devices operating in the UV, because direct transition emission can be adjusted between 3.4eV (GaN) and 6.2eV (AlN). The wide transition range of AlGaN covers the entire lasing wavelength range covered by UV gas and solid state lasers, for example, XeCl(308nm) and KrF(248nm) excimer lasers or N_2(337nm), He-Cd(325nm), SHG-Ar(257nm) lasers. UV semiconductor lasers are attractive in comparison with gas lasers because of small size, long lifetime, high efficiency and continuous-wave (CW) operation. CW-UV lasers using AlGaN materials are believed to replace UV gas lasers in the near future. For the realization of UV semiconductor lasers, several technical problems such as current injection through high Al content AlGaN crystals or efficient UV emission from AlGaN QW structures must be solved. Especially, the realization of high optical gain from AlGaN in UV emission range is most important for the use as the active region of UV lasers.

Many research groups have obtained a strong emission of InGaN QWs when the In content is 10-20%. However the emission efficiency of binary GaN is much weaker than that of InGaN and actually not useful for active region in a semiconductor laser. The mechanism of the efficient emission in InGaN alloy in comparison with GaN has been investigated using Nichia’s samples [4,5]. It was reported that the quantum efficiency of InGaN-based quantum well lasers is enhanced by the effect of localized excitons in nano-scale In segregated (In-rich) regions of the quantum well [5]. The efficient emission of InGaN is observed even when the In incorporation is only a few percent.

On the other hand, AlGaN QW structure with respect to strong UV emission has not yet been well investigated and its optical property is still unknown even though it is very important material in order to realize UV (especially for wavelength shorter than 360nm) optical devices. Recently, we fabricated AlGaN QW and quantum dot (QD) structures for the purpose of intense UV emission [6].

In this work, we report on the $Al_xGa_{1-x}N/Al_yGa_{1-y}N$ (x=0.24-0.53, y=0.11) MQW structures fabricated by metal-organic chemical-vapor-deposition (MOCVD) and demonstrate an intense UV (280-330nm) photoluminescence (PL). We also compare the PL intensity between AlGaN, GaN and InGaN QW structures.

EXPERIMENTS AND DISCUSSIONS

The structures were grown at 76 torr on the Si-face of an on-axis 6H-SiC(0001) substrate, by a conventional horizontal-type MOCVD system. As precursors ammonia (NH_3), trimethylaluminum (TMAl), trimethylgallium (TMGa), and tetraethylsilane (TESi) were used with H_2 as carrier gas. N_2 gas was independently supplied by a separate line and mixed with the H_2 just before the substrate susceptor. Typical gas flows were 2 standard liters per minute (SLM), 2 SLM, and 0.5 SLM for NH_3, H_2, and N_2, respectively. The molar fluxes of TMGa and TMAl for the growth of $Al_xGa_{1-x}N$ (x=0.11-0.53) were 38μmol/min and 2.6-45μmol/min, respectively. At this condition, the growth rate was approximately 2.5μm/h. The substrate temperature measured with a thermocouple located at the substrate susceptor during the growth was 1140°C for all layer.

At first, we investigated the growth condition of high Al content AlGaN alloy. Figure 1 shows the 77K PL spectra of AlGaN films grown directory on very thin (~10nm) AlN buffer layer. The thickness of all the AlGaN film was approximately 400nm. As seen in Fig. 1, single peak emission was obtained for Al contents of 0.11-0.53. The phonon-replica peaks, seen at the low energy side of each spectra, confirms the good crystal quality of the AlGaN. The typical value of full width at half maximum (FWHM) of the spectrum was 20meV for $Al_xGa_{1-x}N$ (x=0.10-0.12) and 65meV for $Al_xGa_{1-x}N$ (x=0.30-0.60) at 77K. For an Al content of 0.6, an additional emission peak around the wavelength of 290nm probably originating from defects was observed besides the 264nm peak. Therefore, the highest Al content we used in this experiment was 0.53.

Figure 2 shows schematic layer structure of the fabricated (a) $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ and (b) $Al_{0.24}Ga_{0.76}N/Al_{0.11}Ga_{0.89}N$ MQW sample. In order to achieve a flat surface suitable for the growth of AlGaN quantum well, an approximately 400nm thick $Al_{0.53}Ga_{0.47}N$ buffer layer for sample (a), and 100nm-thick $Al_{0.24}Ga_{0.76}N$ followed by an 300nm-thick $Al_{0.35}Ga_{0.65}N$ buffer layer for sample (b) were deposited. The buffer layer was found to provide a step-flow grown surface as confirmed by atomic force microscopy (AFM). After that, for sample (a), 5-layer MQW structure consisting of 2.7-6.7nm-thick $Al_{0.11}Ga_{0.89}N$ wells and 8nm-thick undoped $Al_{0.53}Ga_{0.47}N$ barrier layers, and 20nm-thick $Al_{0.53}Ga_{0.47}N$ capping layer were grown. Also, for sample (b), 6-layer MQW structure consisting of 2-5nm-thick Si-doped (undoped) $Al_{0.11}Ga_{0.89}N$ wells and 6nm-thick undoped $Al_{0.24}Ga_{0.76}N$ barrier layers, and 10nm-thick $Al_{0.24}Ga_{0.76}N$ capping layer were grown. The well and barrier thickness is estimated simply from the growth rate of bulk.

Figure 3 shows PL spectra measured at 77K of the undoped $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ 5-layer MQW structures excited with an Ar-SHG laser (257nm) for various well thickness. The spectra of the $Al_{0.53}Ga_{0.47}N$ bulk without $Al_{0.11}Ga_{0.89}N$ well is also shown for comparison of emission intensity. As seen in Fig. 3, the peak wavelength of QW emission shifts from 344nm to 271nm as the well thickness decreases from 6.7nm to 2.7nm. We attribute these shift to the increased quantization energy in the QWs. We cannot see the emission from barrier or capping layers for each MQW spectrum, which indicates that the emission from the quantum well is efficient. The well emission intensity is strongest for a well thickness of 3.3nm. The emission peak wavelength of 400nm-thick $Al_{0.53}Ga_{0.47}N$ buffer layer from the sample without QW is slightly longer than that of 2.7nm-thick MQW structures. This may be because that the barrier bandgap is extended due to the strain compensation between barrier and well. The rapid reduction of the PL intensity with the increase of the well thickness may be caused by a reduction of the radiative recombination probability due to a separation of electron and hole wave-functions in the large piezoelectric field of the well [7]. The reduction of the emission intensity for the thin well may be mainly due to the increase of nonradiative recombination on the hetero-interfaces between well and barrier.

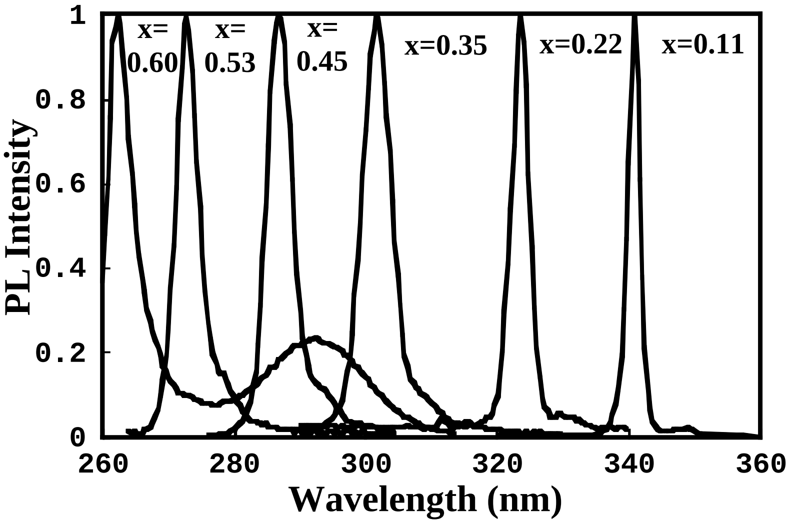

Fig. 1. 77K PL spectra of $Al_xGa_{1-x}N$ (x=0.11-0.60) films grown on the 6H-SiC substrates.

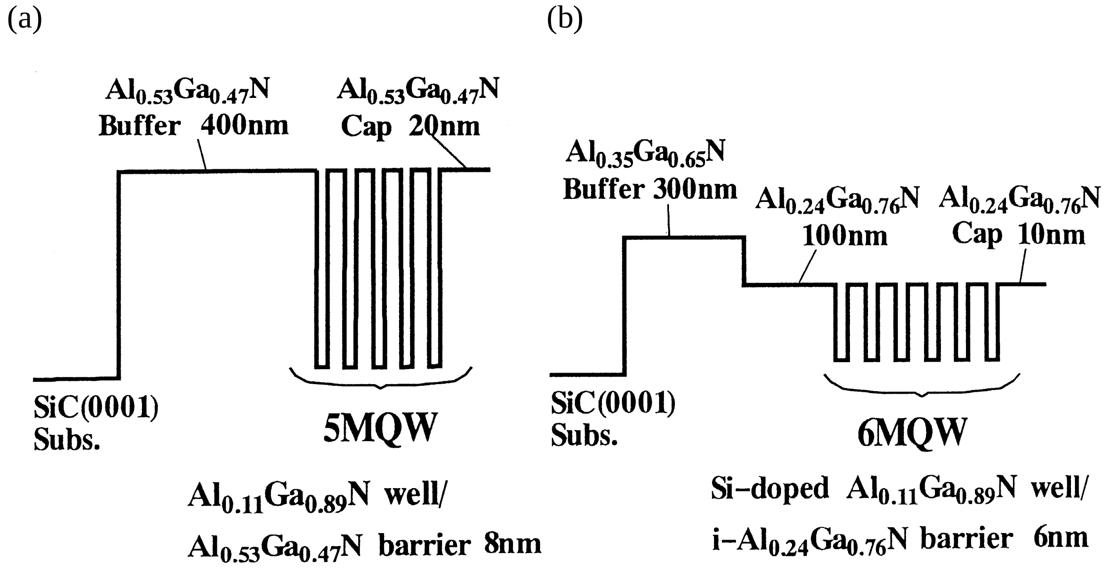

Fig. 2. Schematic layer structure of (a) $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ and (b) $Al_{0.24}Ga_{0.76}N/Al_{0.11}Ga_{0.89}N$ MQW sample.

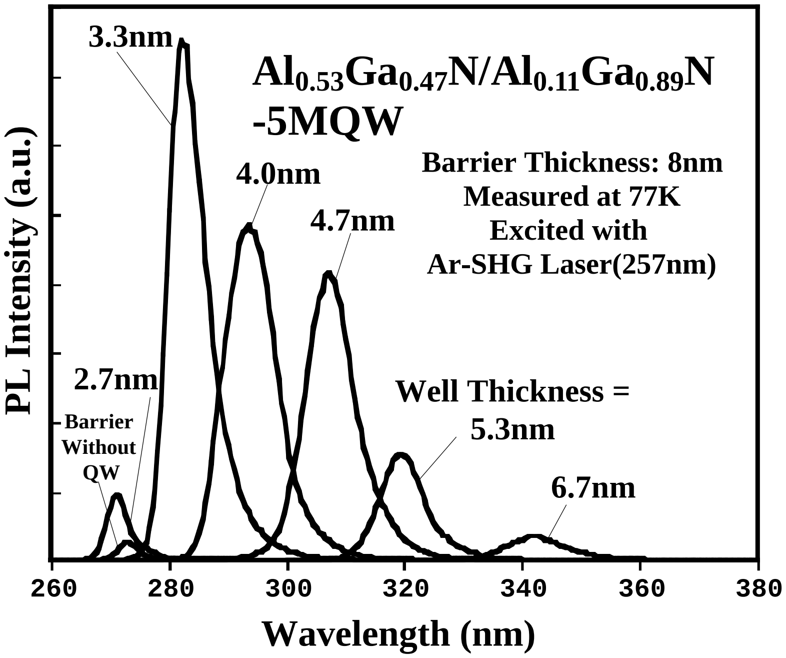

Fig. 3. PL spectra measured at 77K of the undoped $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ 5-layer MQW structures excited with Ar-SHG laser (257nm) for various well thickness.

Figure 4 shows the room temperature PL spectra of the undoped $Al_{0.24}Ga_{0.76}N/Al_{0.11}Ga_{0.89}N$ 6-layer MQW structures excited with a XeCl excimer laser for various well thickness. The peak wavelength of QW emission shifts from 344nm to 323nm as the well thickness decreases from 5nm to 2nm. The 321nm weak emission originates from $Al_{0.24}Ga_{0.76}N$ barrier layers. The dependence of the PL intensity on the QW thickness was similar to that obtained for $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ MQWs. The optimized well thickness was also around 3nm.

Figure 5 shows PL spectra measured at 77K from 3nm thick Si-doped and undoped $Al_{0.24}Ga_{0.76}N$ /$Al_{0.11}Ga_{0.89}N$ 6-layer MQWs excited with a He-Cd laser. Si-doping was used only in the QW layers. The doping concentration was changed from 8×10^{18} to $5\times10^{19}cm^{-3}$. The emission intensity increases drastically by Si-doping. The PL intensity is enhanced by 2-3 times with a Si-doping concentration of $2\times10^{19}cm^{-3}$, as seen in Fig 5. We can see phonon replica peaks on the low energy side of main peaks for a doping concentration below $2\times10^{19}cm^{-3}$. We assume that the screening of piezoelectric field with doping is causing the PL intensity enhancement, as reported in the case of InGaN QWs[8].

Finally, we will compare the emission intensity of AlGaN, GaN and InGaN QWs. Figure 6 shows the PL emission spectra measured at 77K of $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ 5-layer MQW, $Al_{0.24}Ga_{0.76}N/Al_{0.11}Ga_{0.89}N$ 6-layer MQW, $Al_{0.12}Ga_{0.88}N/GaN$ 5-layer MQW and $In_{0.02}Ga_{0.98}N/In_{0.20}Ga_{0.80}N$ single-QW structures. All samples are undoped with optimized well thickness. All samples were excited with Ar-SHG laser with the same excitation condition. As can be seen in Fig. 6, the 280nm emission of the $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ MQW is as strong as that of the $In_{0.02}Ga_{0.98}N/In_{0.20}Ga_{0.80}N$ QW and much stronger than those of the $Al_{0.24}Ga_{0.76}N/Al_{0.11}Ga_{0.89}N$ or $Al_{0.12}Ga_{0.88}N/GaN$ MQWs at 77K. However, the temperature dependence of PL intensity was strongest for $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ MQW and the emission intensity was much reduced at room temperature. We believe that the emission mechanism of AlGaN QWs is much different from that of InGaN QWs. We suggest that the strong UV emission from AlGaN QWs originates in the confinement states which is stable only at low temperature, though, at this moment we don't know the exact mechanism.

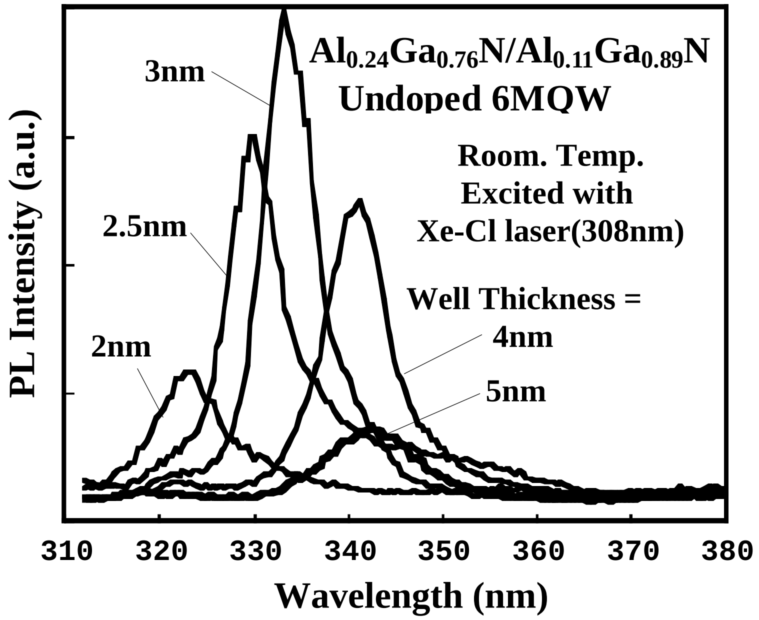

Fig. 4. Room temperature PL spectra for various well thickness from the undoped $Al_{0.24}Ga_{0.76}N/Al_{0.11}Ga_{0.89}N$ 6-layer MQW structures excited with a XeCl excimer laser.

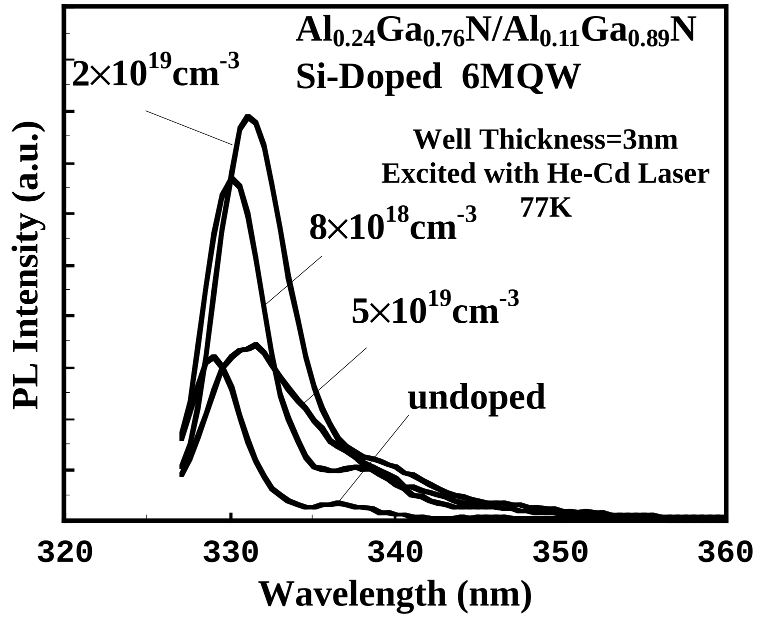

Fig. 5. PL spectra measured at 77K from 3nm thick Si-doped and undoped $Al_{0.24}Ga_{0.76}N$ /$Al_{0.11}Ga_{0.89}N$ 6-layer MQWs excited with a He-Cd laser.

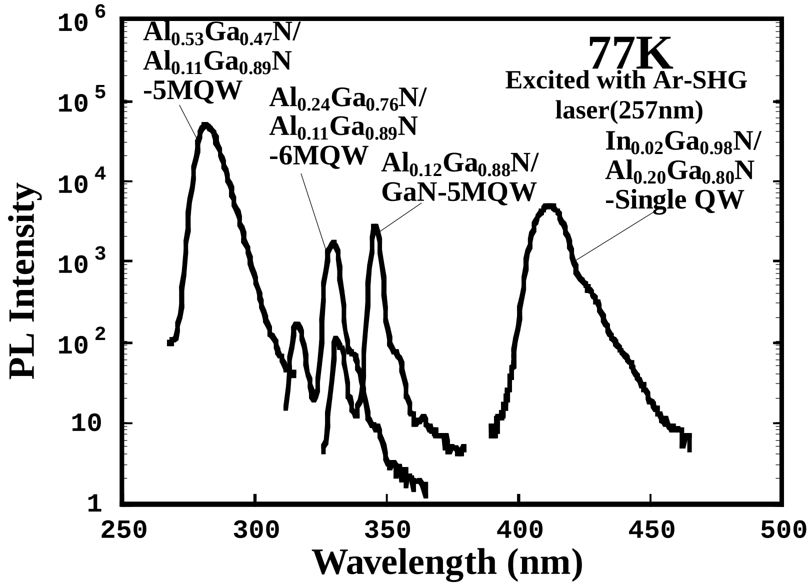

Fig. 6. PL emission spectra of $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$, $Al_{0.24}Ga_{0.76}N/Al_{0.11}Ga_{0.89}N$, $Al_{0.12}Ga_{0.88}N$/GaN MQW and $In_{0.02}Ga_{0.98}N/In_{0.20}Ga_{0.80}N$ single-QW structures.

CONCLUSION

We have demonstrated strong UV (280-330nm) PL emission from AlGaN MQW structures fabricated by MOCVD. Si-doping was shown to be very effective in order to enhance the emission of AlGaN QWs. We investigated systematically the optimized well thickness and the Si-doping concentration of $Al_xGa_{1-x}N/Al_yGa_{1-y}N$ (x=0.24-0.53, y=0.11) MQW structure with respect to efficient emission and found that the optimum values were approximately 3nm and $2\times10^{19}cm^{-3}$, respectively. The, PL intensities of AlGaN, GaN and InGaN quantum well structures were compared. We found that the emission at 77K from a $Al_{0.53}Ga_{0.47}N/Al_{0.11}Ga_{0.89}N$ MQW was as strong as that from the InGaN QWs.

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