Molecular Beam Epitaxial Growth of AlN/GaN Multiple Quantum Wells

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ABSTRACT

AlN/GaN multiple quantum wells (MQWs) were grown on sapphire substrates by plasma-assisted molecular beam epitaxy. Growth temperature, III/V ratio, growth rate, and other growth parameters were optimized for the buffer layer and the MQWs, separately. The growth of AlN buffer was kept as Al-rich as possible while the formation of Al droplets was avoided. A GaN buffer layer was also tried but proved to be inferior to AlN buffer probably due to its larger surface roughness, higher dislocation density, and larger lattice mismatch with the AlN barrier layers in the MQWs. Very flat surfaces with a RMS roughness of 0.7nm were observed by atomic force microscopy (AFM) on the samples with both AlN buffer layer and 20 MQWs deposited under the optimized growth conditions. Abrupt interfaces and excellent periodicities of the MQWs were confirmed by X-ray diffraction (XRD) and reflectivity measurements with MQWs' satellite peaks clearly visible up to the 10th order. Room-temperature intense ultraviolet (UV) photoluminescence (PL) emission with wavelength in the range of 320-350nm was also observed from the MQWs with well width ranging from 1.0 to 1.5nm. These MQW structures can potentially be used for UV light emitters and quantum cascade lasers.

INTRODUCTION

There have been considerable interests in group III-nitrides for their applications in the ultraviolet (UV) light emitter diodes (LEDs) and laser diodes (LDs) [1-3]. Until recently, most of the efforts had been focused on InGaN-based light emitters [1,2]. But to achieve light emission with even shorter wavelength, AlGaN-based light emitting structures need to be used. Due to the fact that low-dimensional heterostructures can provide carrier confinement and therefore

improve the optical efficiency, AlGaN/GaN quantum wells with low aluminum concentration AlGaN barriers have been used to make UV LEDs [4]. In order to further shorten the emission wavelength, higher aluminum concentration AlGaN or even AlN barriers should be used in the quantum well structures. Besides the applications using intraband transitions, AlGaN/GaN quantum well system is also a potential candidate for quantum cascade lasers due to the recent realization of intersubband transitions (absorption, so far) [5]. High aluminum concentration AlGaN or AlN barriers are also needed in this case to maximize the intersubband transition energy for potential applications in short wavelength quantum cascade lasers.

In this study, we fabricated AlN/GaN MQWs by molecular beam epitaxy and optimized the growth conditions. Strong room-temperature UV PL emission was observed from the MQWs.

EXPERIMENT

The structures used in this study were grown in a Varian Gen II molecular beam epitaxy (MBE) system which used standard effusion cells for the group III elements and an EPI Unibulb rf plasma source to supply nitrogen radicals. The MQWs were grown typically with the rf power and growth chamber pressure at approximately 250 W and 1.5×10^{-5} torr, respectively. All the growths took place on 2 inch c-plane sapphire substrates which were backside coated with TiW for more efficient heating. The wafer pretreatment process included a one-hour outgassing step at 450°C and a following 30-minute low temperature nitridation step at 160°C with the rf power set at 500 W. Growth temperatures were monitored by a pyrometer calibrated to measure the temperature of the TiW on the backside of the substrates.

The epitaxial structures consisted of a buffer layer of 0.3-0.5 μm, which was either AlN or GaN, and 20 AlN/GaN quantum wells on all the samples in this study. Different growth conditions were experimented in an attempt to achieve the best quality of the MQWs. For all the samples studied, the AlN barrier thickness was kept at 60Å, while the thickness of GaN wells varied from 9 to 15Å. The nominal thickness of each layer was calculated by the growth rates of equivalent bulk samples. No growth interruptions occurred during the deposition of MQWs.

## RESULTS AND DISCUSSION

Growth Optimization

The growth optimization process was made by the comparisons of the MQWs' quality among the samples grown under different conditions (see Table I.). The entire epitaxial structure of all the samples listed in Table I was undoped. Also listed in the table is the RMS surface roughness measured by AFM for each sample with both the buffer and MQWs already deposited. None of the samples listed in Table I exhibited cracks on the surface. The first thing to optimize in this study was the material of the buffer layer. In our case, for simplicity, only AlN and GaN buffer layers were tried. The growth conditions we used for the GaN buffer layer were expected to produce the GaN epitaxial layer of the best quality in our MBE system, based on our previous experience. But according to the surface roughness measurements, the MQW sample with GaN buffer (sample "E") had the poorest surface morphology with an RMS roughness of 4.9 nm, which was higher than any of the MQW samples grown on AlN buffer. Besides AFM measurements, the quality of the MQWs on some of these samples was also evaluated by X-ray diffraction and reflectivity measurements. A comparison of X-ray diffraction pattern was made

Table I. Structures of the samples for growth optimization and the measured surface roughness.

Sample	Buffer	Growth temperature of buffer (°C)	Growth temperature of MQWs (°C)	Nominal thickness of GaN wells (Å)	Surface RMS roughness (nm)
A	AlN	700	700	10	2.8
B	AlN	700	700	14	3.3
C	AlN	800	700	12	0.7
D	AlN	800	800	14	3.2
E	GaN	800	800	14	4.9
F	AlN	800	700	14	1.3

between sample "E" and sample "D", which had the same structures and growth conditions except for the buffer. The X-ray pattern of sample "E" showed almost no satellite peaks of MQWs, while satellite peaks up to the 3rd order were clearly resolved in the case of sample "D", which indicated that the MQWs on sample "D" had more abrupt interfaces, better periodicities, and probably a flatter surface of buffer layer to start with. This, along with the AFM results, proved that the GaN buffer was probably inferior to the AlN buffer for the growth of AlN/GaN MQWs possibly due to its larger surface roughness, higher dislocation density, and larger lattice mismatch with the AlN barrier layers in the MQWs. The second parameter optimized was the growth temperature of the AlN buffer layer. Based on the surface roughness results, the samples with the high temperature (800°C) buffer ("C" and "F") had consistently much smoother surface than the samples with low temperature (700°C) buffer ("A" and "B"), if the MQWs were grown at the same temperature (700°C in this study). This suggested that a high temperature AlN buffer should be a preferred candidate for the growth of AlN/GaN MQWs due to its flatter surface and possibly lower dislocation density. Both X-ray diffraction and reflectivity measurements confirmed this finding. The X-ray diffraction profile of sample "C" exhibited more and also much stronger satellite peaks than that of sample "A" or "B", while the X-ray reflectivity measurements gave similar results on sample "C" and "B" (see Figure 1.). The next very
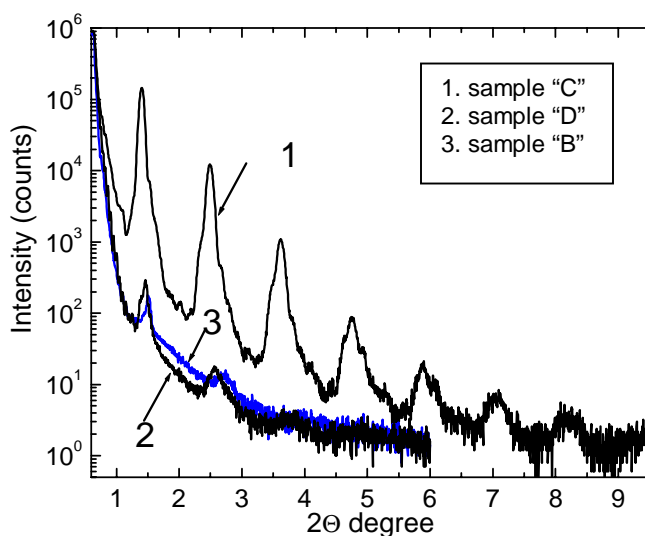
Figure 1. X-ray reflectivity measurements of AlN/GaN MQW samples.

important growth parameter was the growth temperature of the AlN/GaN MQWs. The results of AFM (sample "C", "D", and "F" in Table I.), X-ray diffraction (much more and stronger satellite peaks on sample "C" and "F" than on sample "D"), and X-ray reflectivity (sample "C" and "D" in Figure 1.) measurements all pointed to the same conclusion that AlN/GaN MQWs grown at low temperature (700°C) had much better quality and flatter surface than those grown at high temperature (800°C). One possible explanation is the growth mode dependence on the substrate temperature when GaN is grown on AlN [6]. However, so far, no solid evidence has been observed in reflection high energy electron diffraction (RHEED) or other measurements to support this explanation. Further study is required. Another observation worth mentioning is that according to the data in Table I, the thickness of the GaN wells did not seem to affect as much the surface roughness as the growth temperatures or the buffer material.

In order to have more precise control of the thickness of MQW layers, we grew the MQWs at a growth rate of 0.3-0.4μm per hour, instead of 0.5-0.6μm per hour which is typical and probably optimum (to get the best material qualities, based on our previous experiences) for the buffer layer growth. This was done by using relatively lower group-III fluxes and rf power of nitrogen plasma during the MQW growth. It is not clear at this point whether such a reduced growth rate is optimum or not for the MQWs in terms of material qualities.

Apart from the growth parameters mentioned above, we also found that during the AlN buffer growth, the more Al-rich the III/V ratio was, the better the AlN quality we obtained. In order to make the growth as Al-rich as possible without forming Al droplets, we adjusted the III/V ratio so that the 2" wafer was covered by Al droplets except for the central area about 1" in diameter due to the radial ununiformity of the plasma density and substrate heating efficiency. In doing so, the AlN buffer layer grown in the central area of the wafer usually had flatter surface and lower dislocation density than those grown without using this method.

Optimizedly-grown MQWs

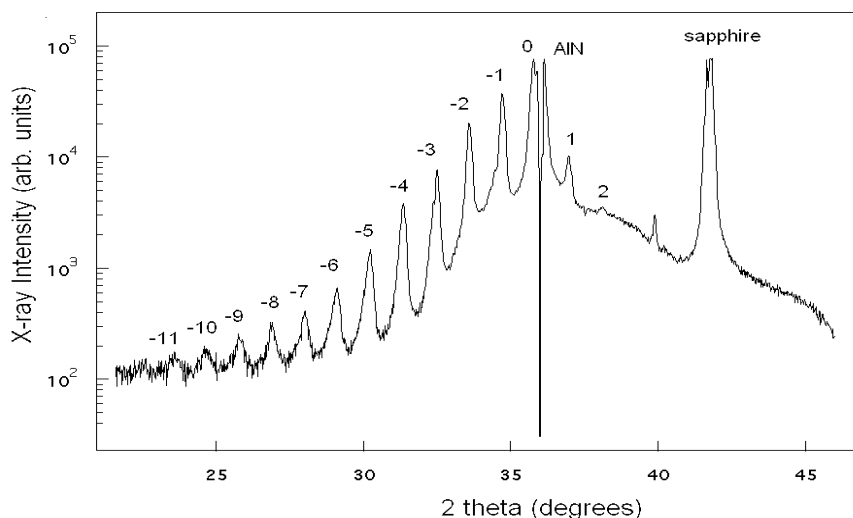A series of AlN/GaN MQWs with different well thickness were grown under the optimized

Figure 2. X-ray diffraction profile (θ–2θ scan) of AlN/GaN MQWs. The kinks (sharp decrease in signal) at the peaks of AlN and sapphire were due to the saturation of X-ray detector.

conditions which were used in the growths of sample "C" and "F" listed in Table I. A typical X-ray diffraction pattern of these MQWs is shown in Figure 2. Well resolved satellite peaks up the 10th order were observed, which indicated that abrupt interfaces and excellent periodicities of the MQWs were achieved. Surface RMS roughness of 1.0nm or smaller was obtained on these samples. The high quality of MQWs was also confirmed by transmission electron microscopy (TEM) measurements. In Figure 3, the TEM image exhibits near monolayer-abrupt interfaces between GaN wells and AlN barriers, considering the fact that the quantum wells, except the first one, shown in the figure were only approximately 5-monolayer-thick. It is interesting to note that the first quantum well was unexpectedly thin, which was probably due to the excess Al from the Al-rich buffer forming high Al concentration AlGaN by competing with Ga to react with nitrogen radicals when the growth of the first GaN well started. This could have been avoided if the wafer had been exposed to the nitrogen plasma for a short period of time immediately after the AlN buffer growth was finished. Besides, the comparisons between the simulated X-ray diffraction profile (not shown in Figure 2.) and the measured X-ray data showed that the AlN barriers were almost fully relaxed while the GaN wells were almost fully strained, which was expected due to the relatively thick AlN buffer layer.

Finally, room-temperature PL measurements were made on these AlN/GaN MQW samples grown with the optimized conditions [7]. Single-peak strong UV emission with the peak energy in the range of 3.45-3.97eV was observed from the samples with the well width ranging from 0.9 to 1.5nm (see Figure 4.). The well width dependence of the peak energy agrees fairly well with the theoretical calculation when the piezoelectric and spontaneous polarization effects are taken into account.
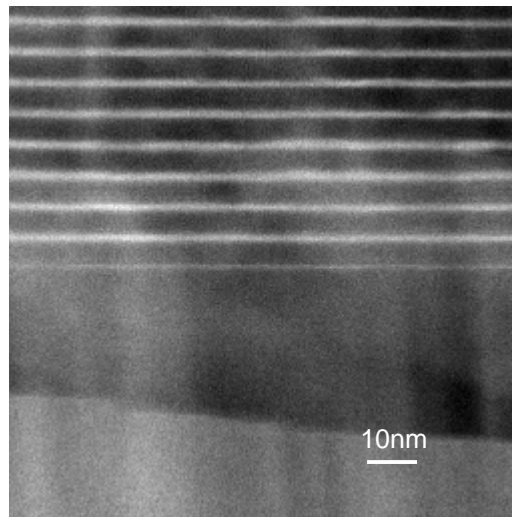

Figure 3. TEM image (dark field) of AlN/GaN MQWs. The first 9 quantum wells are shown. The brighter layers are GaN wells and the rest is AlN. The nominal thickness of GaN wells and AlN barriers on this sample is 1.3 and 6.0nm, respectively. The abrupt color change near the bottom of the image was due to the fact that a piece of sample peeled off during TEM sample preparation.
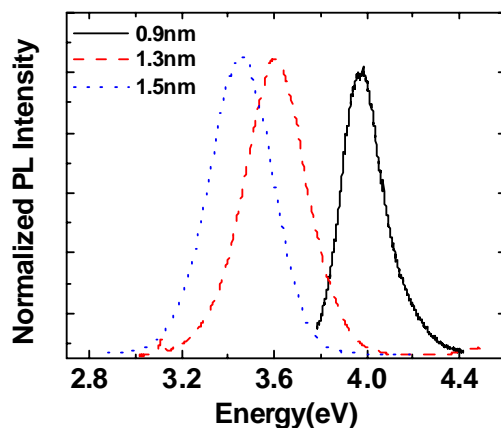
Figure 4. Room temperature time-integrated photoluminescence spectra from three AlN/GaN MQW samples with the well thickness of 0.9, 1.3, and 1.5nm, respectively.

CONCLUSIONS

This study shows the optimization process of the molecular beam epitaxial growth of AlN/GaN MQWs. Grown under the optimum conditions, the MQWs showed near monolayer-abrupt interfaces and coherent periodicities. Room temperature strong UV emission was observed from these MQWs.

ACKNOWLEDGEMENTS

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