AlGaN/GaN MULTIPLE QUANTUM WELLS GROWN BY ATOMIC LAYER DEPOSITION

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ABSTRACT

A three-pair AlGaN/GaN multiple quantum well (MQW) structure with superlattices (SLs) was grown on c-plane sapphire using metal organic chemical vapor deposition (MOCVD) system. The AlGaN barrier and GaN well of the MQW structure were grown by atomic layer deposition (ALD) and conventional growth, respectively. The HRTEM and HRXRD results show the grown structure has shape interface between SLs layers and QWs with good periodicity. The AFM and SEM data show smooth surface morphology with low RMS value and low defect density. The CL measurements also indicate uniform luminescence pattern at room temperature. The AlGaN/GaN MQW with AlN/GaN SLs structure grown by ALD could be used to improve the surface morphology by effectively suppress the threading dislocation.

Keywords: Atomic layer deposition, AlGaN, UV, quantum well

1. INTRODUCTION

The III-nitride based semiconductors have become the most important materials for light-emitting diodes (LEDs) and laser diodes (LDs) under continuous wave operation in the visible spectrum region ranging from green to blue colors [1-4]. Recently, the ultraviolet LEDs (UV-LEDs) have attracted much attention because it shows a potential for developing white light LEDs. Because the phosphor based white LEDs excited by the UV-LEDs has higher efficiency than that of white LEDs excited by the blue LEDs and YGA phosphor. Besides, the UV-LEDs have many potential applications such as optical data storage, microscopes, and lithography instruments. Therefore AlGaN-based materials are suitable candidate for development of ultraviolet light source. However, it is still difficult to grow device-quality AlGaN-based epilayer on sapphire substrate due to the large lattice mismatch and the thermal expansion coefficient incompatibility. In addition, the AlGaN/GaN MQWs is very sensitive to the dislocation and defects that are produced by the lattice mismatch between GaN and the sapphire substrate. Furthermore unlike the blue/green InGaN/GaN MQWs which have a strong localization center to effectively keep carriers away from nonradiative pathways and to provide a radiative channel for enhancement of the PL quantum efficiency [5]. To develop an efficient UV LED based on the AlGaN/GaN MQWs still have much problem to solve. In particular the surface morphology of AlGaN/GaN MQWs and threading dislocation density play an important role in optical properties of LED devices. Recently the relatively high quality AlGaN/GaN heterostructures using quasi AlGaN as a barrier layer which is formed by AlN/GaN super-lattice was reported [6-7]. In this paper, we report growth of high quality AlGaN/GaN quantum well structure using the atomic layer deposition (ALD) AlN/GaN SLs as AlGaN barrier. The grown sample structure showed shape interface between SLs layers and QWs with good periodicity. The AFM and SEM data show smooth surface morphology with low RMS value and low defect density. The optical properties were also experimentally characterized by PL and CL measurements.

2. EXPERIMENT

The AlGaN/GaN MQW structures were grown by commercial low-pressure metalorganic chemical vapor deposition (VEECO D75 system). The liquid MO compounds of TMGa, TMAl and gaseous NH_3 were employed as the reactant source materials for Ga, Al and N, respectively and H_2 and N_2 were used as the carrier

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gas. The substrates used in this experiment were cut from (0002)-oriented sapphire wafers offset by 0.2° off toward <01-11> direction. The AlGaN barrier and GaN well of the MQW structure were grown by ALD and conventional growth, respectively. The ALD process involves alternate control of mass flow of trimethylaluminum and trimethylgallium gas during the growth of AlGaN barrier to form superlattice (SLs). The MO gas flow time of AlN and GaN layer were 6 and 19 sec respectively under a continuous flush of the NH_3 gas at 850°C. The short interruption time is 10 sec between AlN and GaN. Under these growth conditions, the grown rate of ALD AlGaN is 0.14 µm which is lower than the conventional growth rate of about 0.6 µm/hr~1 µm/hr [8]. The whole structure consists of three GaN QWs and three AlGaN barriers as shown in Fig. 1. The surface morphology of the top layer was observed by atomic force microscopy (AFM). Crystalline quality was evaluated by high-resolution x-ray diffraction (HRXRD) and Cu K_α radiation was used as the X-ray source. The average thicknesses of the AlGaN barriers and the GaN wells were determined from the angular distance using satellite peaks of $\omega/2\theta$-scan diffraction patterns. A transmission electron microscope (TEM) was used to study thickness and the sharpness of the AlGaN/GaN structure interfaces. For the photoluminescence (PL) measurements 266 nm pulse laser was used as excitation source. The pulse width is 200 fs and the repetition rate is 76 MHz. The maximum average power is 20 mW. And the luminescence signal dispersed through a 0.5-meter monochromator was detected by photomultiplier tube (PMT). CL measurements were carried out at 300 K by using a MonoCL system installed on a field emission scanning electron microscope with beam energies of 5–20 keV.

3. RESULTS AND DISCUSSION

Fig. 1 (a) shows a cross-sectional HRTEM image of three pairs AlGaN/GaN quantum well structure. The insert graph in Fig. 1 shows the enlarged MQWs schema and the scale bar is 5 nm. Based on the Fig. 1(a) results, the ALD grown AlGaN barrier consists of six pairs of AlN/GaN superlattices with AlN thickness of 4.3 Å and GaN thickness of 7.7 Å respectively to form a AlGaN barrier thickness of 7.2 nm, while the GaN well has a thickness of 3 nm. The bright parts and dark parts correspond to the AlN and the GaN, respectively. The results show a clear image of interface and mono-atomic step indicating successful formation of superlattice by ALD process. Note that the interfaces of the AlGaN barriers on the GaN wells are as sharp as those of the GaN wells on the AlGaN barriers. On other hand, no threading dislocations running across the sample are seen in Fig. 1(a). Moreover, we also have observed the suppression of threading dislocation under bottom barrier layer with SLs in Fig. 1(b). The ALD grown AlGaN barrier layer can reduce the threading dislocation. The HRTEM images also show suppression of treading dislocation by the SLs suggesting the AlGaN barrier with SLs could be used to improve surface morphology and reduction of defect density of the MQWs [8]. We also don't see the any crack on the wafer surface.

The HRXRD pattern of the $\omega/2\theta$-scan (0002) reflections for AlGaN/GaN MQWs structure is shown in Fig. 2. The diffraction pattern shows the two periodical structures. One is the AlGaN/GaN MQWs, another is AlN/GaN SLs in the barrier of quantum well. The solid and dashed lines show the experimental and simulated results, respectively. The high order satellite peaks of QWs and SLs are clearly observed shown in Fig. 2(a). The five satellite peaks of QWs are from -3th to 2th. We also observed the -1 order peak of SLs. The separation of QWs and SLs satellite peak is 1.6 K and 9.0 K arcsec. These separations exhibit the thickness of periodical structure which indicates smooth and abrupt interfaces with good periodicity of the SLs and QWs structure. According to the simulation results, the six pairs of SLs structure have a thickness of 0.42 nm and 0.77 nm for AlN and GaN respectively forming a AlGaN barrier layer with a overall thickness of 7.14 nm. While the GaN well has a thickness of 2.9 nm. The result was similar to our HRTEM data in inserting graph of FiG. 1(a). The reciprocal space mapping (RSM) of the grown structure was obtained as shown in Fig. 2 (b). The RSM data show asymmetry (1 0 -1 5) reflection for the grown sample with clear AlGaN and GaN bulk peak observed in Fig. 2(b). The RSM results show the same the in-plane lattice constants as the underlying GaN layer. It indicates that the AlGaN layer to be full strained and pseudomorphic to the underlying GaN layer [9]. This result also suggested the relative tensile stress in the AlN layer in the SLs was reduced and the crack generation could be suppressed. The AFM measurement showed the root-mean-square surface roughness is about 0.35 nm which is about 2~3 times smoother than those grown by conventional method[6]. Therefore the atomic layer deposition technique we used allows growth of high quality AlGaN/GaN quantum well.

Fig. 4 (a) and (b) shows the PL spectra at different temperature and CL spectrum at room temperature of the sample. In Fig. 4(a) and (b) all spectra exhibited two emission peaks. One is the band edge emission of GaN bulk at 3.43 eV, another emission band is the quantum well GaN between the AlN/GaN SLs as AlGaN barrier. The AlGaN/GaN quantum well emission energy at 13 K and 300 K is 3.77 eV and 3.71 eV, respectively.

The surface morphology from SEM measurement is flat and featureless as shown in Fig. 5(a). Fig. 5 (b) and 5 (c) show the CL images of the bulk GaN and GaN quantum well emission for E= 3.39 eV and E= 3.71 eV, respectively. All images were taken at the same position. There are many clearly observed dark spots in Fig. 5(b). In the contrast, the 5 (c) showed uniform emission intensity without dark spots. From the early report the dark spot of bulk GaN image pattern is related to dislocation [10]. The dark region is indicated the non-radiative recombination center in bulk GaN due to threading dislocation or defects in the material. From these data the surface morphology was improved by using AlN/GaN SLs as an AlGaN barrier layer in the quantum well grown ALD technique.

4. CONCULSIONS

Three pairs AlGaN/GaN MQWs with AlN/GaN SLs as an AlGaN barrier layer was grown by ALD. The HRTEM and HRXRD results show the grown structure has shape interface between SLs layers and QWs with good periodicity. The AFM and SEM data show smooth surface morphology with low RMS value and low defect density. The CL measurements also indicate uniform luminescence pattern at room temperature. The AlGaN/GaN MQW with AlN/GaN SLs structure grown by ALD could be used to improve the surface morphology by effectively suppress the threading dislocation.

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REFERENCES

[1] S. Nakamura, T. Mukai, and M. Senoh, "High-brightness InGaN/AlGaN double-heterostructure blue-green-light-emitting diodes, " J. Appl. Phys. **76** (1994) 8189.

[2] S. Nakamura, M. Senoh, and T. Mukai, "P-GaN/N-InGaN/N-GaN double-heterostructure blue-light-emitting diodes, " Jpn. J. Appl. Phys. **32** (1993) L8.

[3] T. Nishida, H. Saito, and N. Kobayashi, "Milliwatt operation of AlGaN-based single-quantum-well light emitting diode in the ultraviolet region, " Appl. Phys. Lett. **78** (2001) 3927.

[4] C. Q. Chen, J. W. Yang, M.-Y. Ryu, J. P. Zhang, E. Kuokstis, G. Simin, and M. A. Khan, "Pulsed Metalorganic Chemical Vapor Deposition of Quaternary AlInGaN Layers and Multiple Quantum Wells for Ultraviolet Light Emission, " Jpn. J. Appl. Phys. Lett. **41** (2002) 1924.

[5] S. F. Chichibu, A. C. Abare, M. S. Minsky, S. Keller, S. B. Fleischer, J. E. Bowers,E. Hu, U. K. Mishra, L. A. Coldren, and S. P. DenBaars, " Effective band gap inhomogeneity and piezoelectric field in InGaN/GaN multiquantum well structures, " Appl. Phys. Lett. **73** (1998) 2006

[6] Y. Kawakami, X.Q. Shen, G. Piao, M. Shimizu, H. Nakanishi, H. Okumuraa, "Improvements of surface morphology and sheet resistance of AlGaN/GaN HEMT structures using quasi AlGaN barrier layers, " J. Cry. Growth, **300** (2007)168-171.

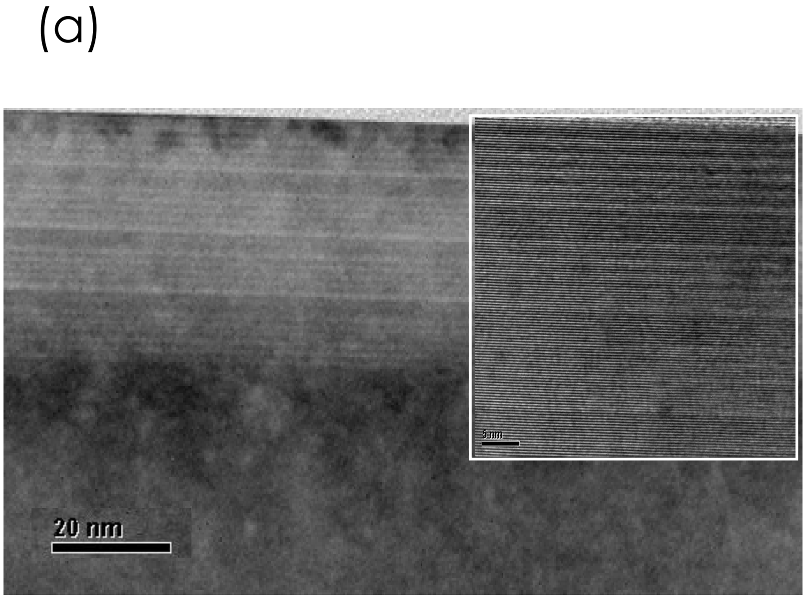
[7] Y. Kawakami, A. Nakajima, X. Q. Shen, G. Piao, M. Shimizu, and H. Okumura, " Improved electrical properties in AlGaN/GaN heterostructures using AlN/GaN superlattice as a quasi-AlGaN barrier, " Appl. Phys. Lett. **90** (2007) 242112

[8] H. Tokunaga, A. Ubukata, Y. Yano, A. Yamaguchi, N. Akutsu, T. Yamasaki, K. Matsumoto, " Effects of growth pressure on AlGaN and Mg-doped GaN grown using multiwafer metal organic vapor phase epitaxy system, " J. Cryst. Growth. **272** (2004) 348.

[9] A. Torabi, W. E. Hoke, J. J. Mosca, J. J. Siddiqui, R. B. Hallock, and T. D. Kennedy, "Influence of AlN nucleation layer on the epitaxy of GaN/AlGaN high electron mobility transistor structure and wafer curvature J. Vac. Sci. Technol. **B 23**(2005) 1194.

[10] Hai Lu, X.A. Cao, S.F. LeBoeuf, H.C. Hong, E.B. Kaminsky, S.D. Arthur, " Cathodoluminescence

mapping and selective etching of defects in bulk GaN, " J. Cryst. Growth, **291** (2006)82-85.



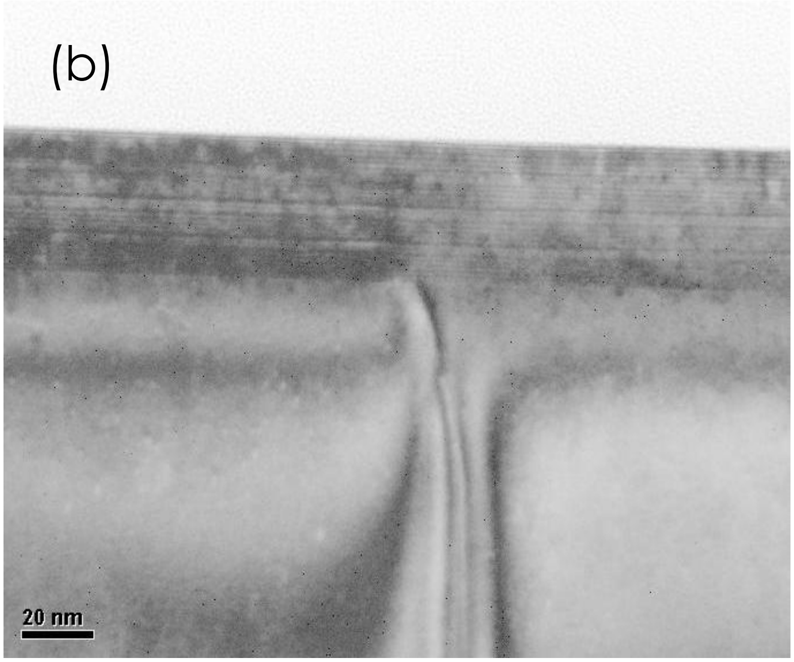


Figure 1(a) Transmission electron microscope image of (a) of the three pairs AlGaN/GaN multiple quantum well structure and the insert image is the part of the MQWs. Fig.1(b) A threading dislocation from the substrate could be suppressed by three pairs AlGaN/GaN multiple quantum well structure.

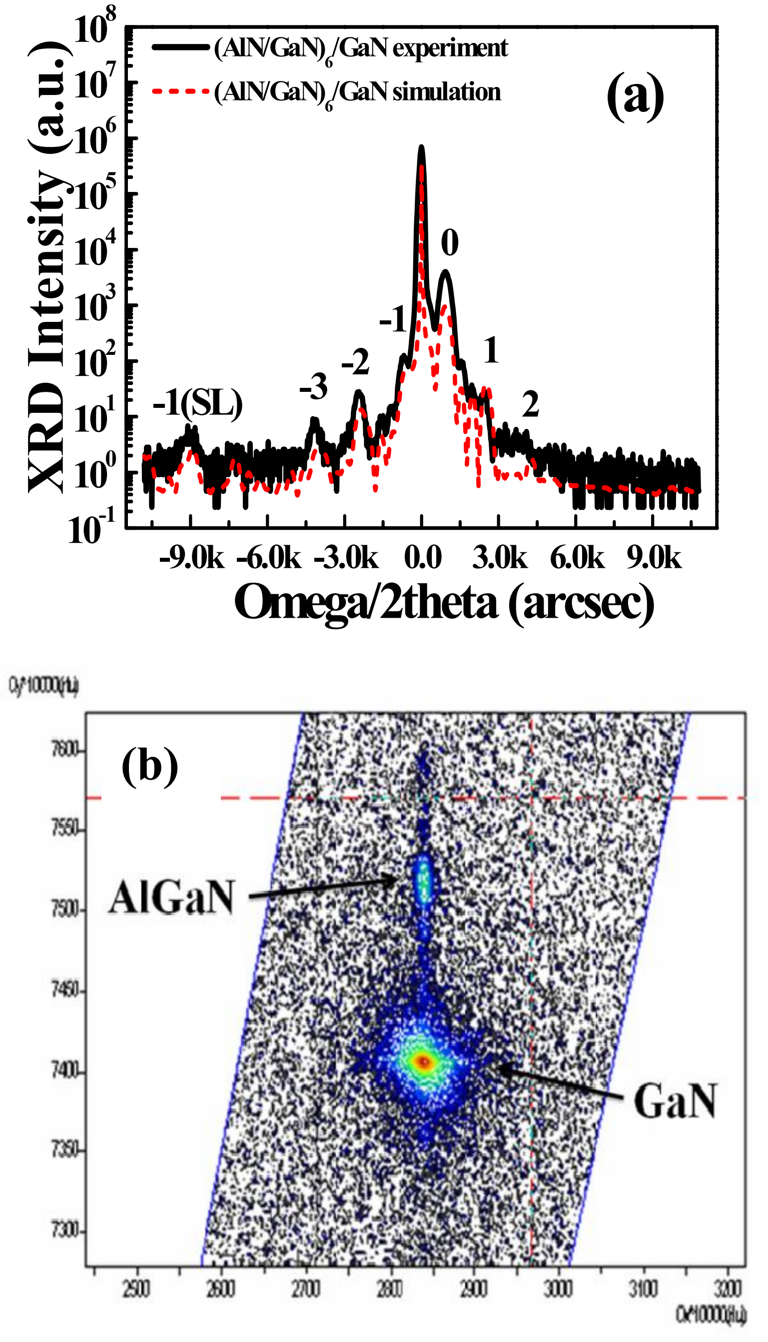


Figure 2 (a) (0 0 0 2) ω-2θ x-ray diffraction pattern and simulated result of AlGaN/GaN MQWs. The satellite peaks of QWs from the -3th to 2th. The -1th satellite peak of SLs is located at the -9.0 K form the GaN peak. The Fig. 2 (b) is show the reciprocal space mapping (RSM) of asymmetry (1 0 -1 5) reflection.

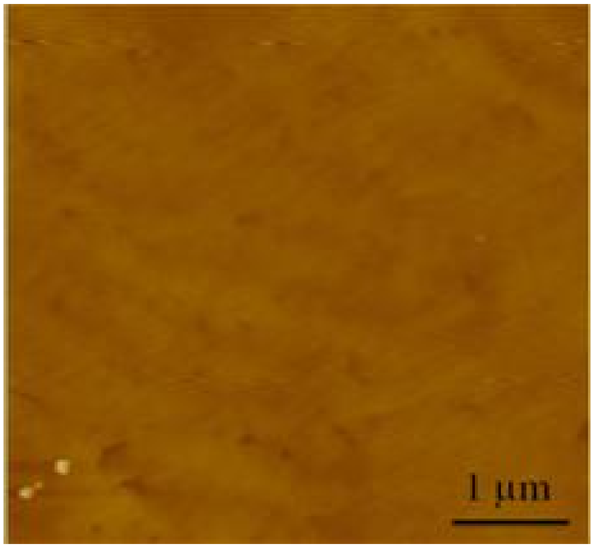


Figure 3 AFM image of AlGaN/GaN MQWs, the root mean square roughness is 0.35 nm. The scan area is 5 x 5 μm^2

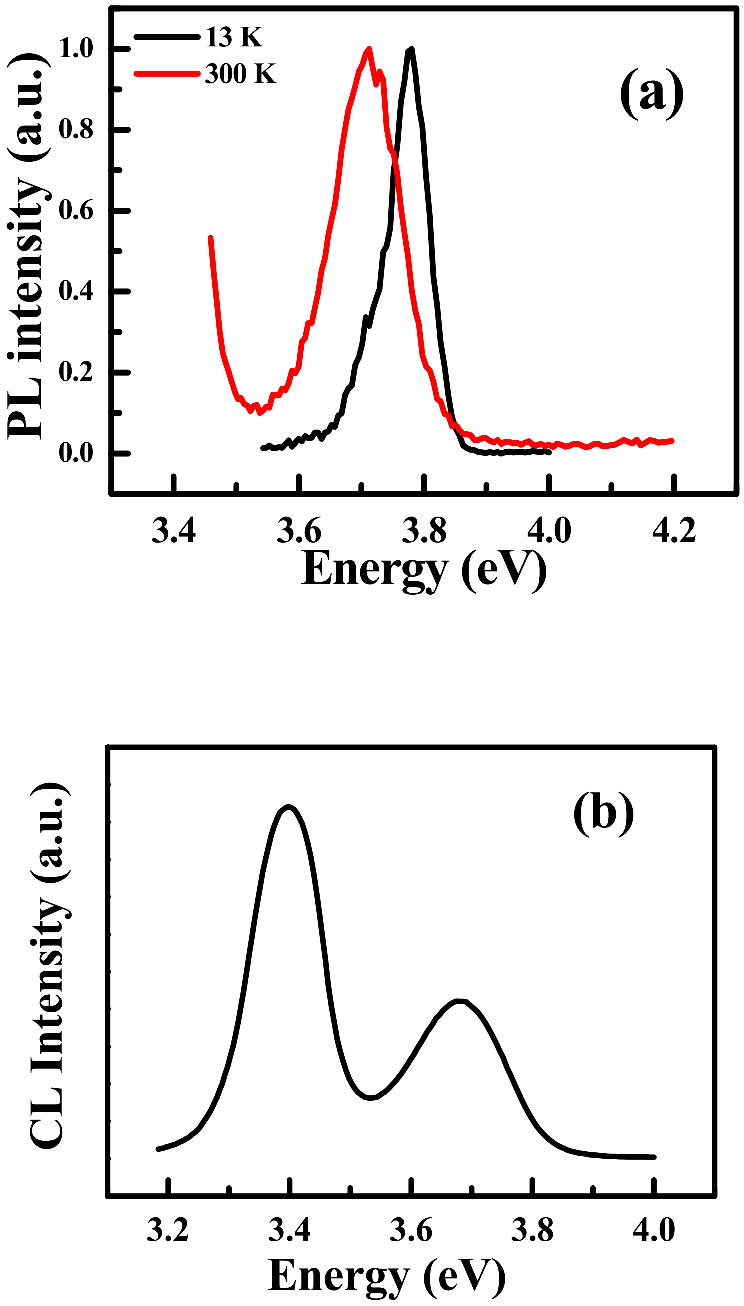


Figure 4 (a) The PL spectra of AlGaN/GaN MQWs at different temperature. Right inset figure shows the PL spectrum at low intensity scale. (b) The CL spectrum at room temperature.

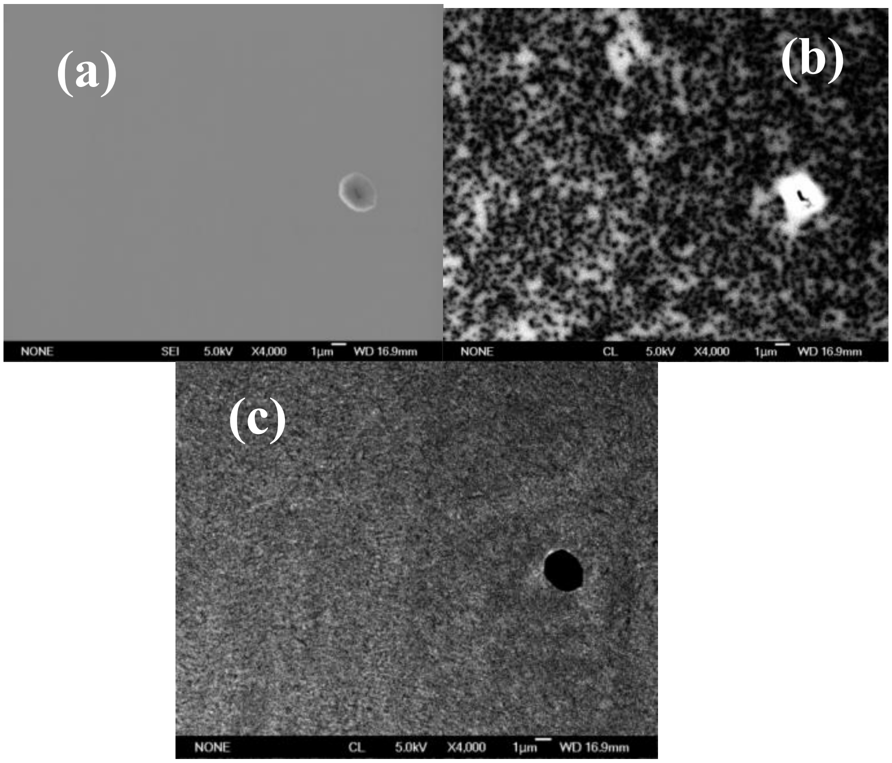


Figure 5 (a) Scan electron microscope image of AlGaN/GaN MQWs sample, (b) CL image taken at E= 3.39 eV and (c) E= 3.71 eV