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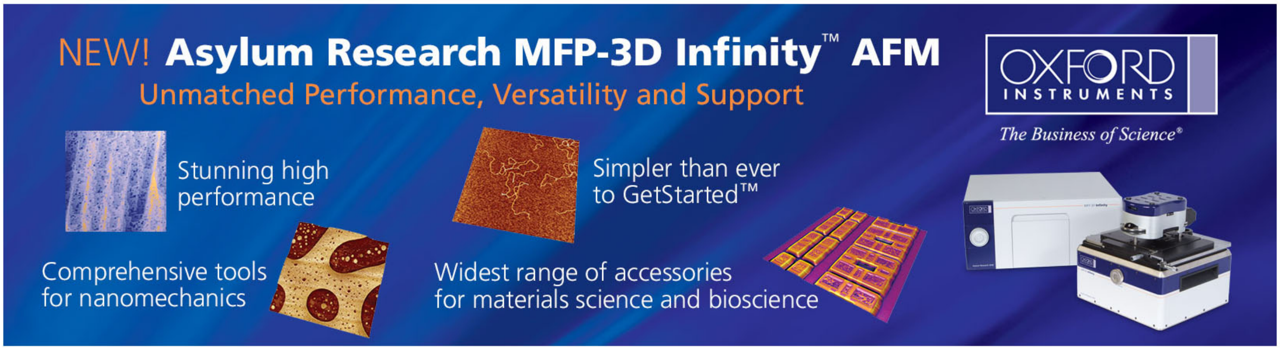

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High quality ultraviolet AlGaN/GaN multiple quantum wells with atomic layer deposition grown AlGaN barriers

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Low dislocation density ultraviolet (UV) AlGaN/GaN multiple quantum well (MQW) structure was grown using atomic layer deposition (ALD) technique. The AlGaN/GaN MQW grown on the sapphire substrate consisted of three GaN QWs and four AlGaN barriers formed by ALD grown AlN/GaN superlattices. The as-grown sample showed smooth surface morphology with a root-mean-square roughness value of only 0.35 nm, and no surface cracks were found. The dislocation density was estimated to be as low as 3.3×10^7 cm^{-2}. X-ray and transmission electron microscope data showed the MQW had sharp interfaces with good periodicity. The sample had an UV photoluminescence emission at 334 nm (3.71 eV) with a very narrow linewidth of 47 meV at 13 K. The cathodoluminescence image revealed a fairly uniform luminescence pattern at room temperature. The AlGaN/GaN MQW grown by ALD technique should be useful for providing high crystalline quality for fabrication of various optical devices. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2996566]

The AlGaN/GaN multiple quantum wells (MQWs) have attracted much attention because of their unique properties, such as a high conduction band offset, better carrier confinement, large longitudinal (LO) phonon energy, and ultrafast carrier and intersubband relaxation, making AlGaN/GaN MQWs promising structures for realizing ultraviolet light emitting diodes and laser diodes.[1–5] Recent reports indicated that the optical and electrical properties of AlGaN/GaN MQWs were very sensitive to the crystalline quality and the threading dislocation density in the AlGaN/GaN epilayer.[6,7] So far most AlGaN/GaN MQWs structures were grown on lattice-mismatched foreign substrates such as sapphire, making it difficult to grow device-quality MQWs due to the lattice mismatch and the misfit in the thermal expansion coefficients between these two material systems. Recently high quality AlGaN/GaN heterostructures using quasi-AlGaN formed by AlN/GaN superlattices (SLs) as barrier layers were reported.[8,9] However, these results mainly focused on the electrical properties used for AlGaN/GaN high electron mobility transistors and no optical properties were reported. In this paper, we report the growth of low dislocation density and crack-free AlGaN/GaN MQWs by using the atomic layer deposition (ALD) grown AlN/GaN SLs as the AlGaN barrier. The as-grown AlGaN/GaN MQWs sample had low defect density, smooth surface morphology with small root-mean-square (rms) roughness value and sharp interfaces. In addition, the AlGaN/GaN MQWs sample showed a sharp photoluminescence (PL) spectrum and a uniform cathodoluminescence (CL) pattern.

The AlGaN/GaN MQW structures were grown by the low-pressure metal-organic chemical vapor deposition VEECO D75 system. The trimethylgallium (TMGa), trimethylaluminium (TMAl), and gaseous NH_3 were employed as the reactant sources for Ga, Al, and N, respectively, and H_2 and N_2 were used as the carrier gaseous. The (0001)-oriented sapphire substrate with a 0.2° offset was first heated to 1000 °C under a H_2 ambient for 5 min. Then, a 2-μm-thick GaN epilayer was grown after the deposition of a low-temperature nucleation layer. Finally, the AlGaN/GaN MQWs structure comprising three GaN wells and four AlGaN barriers were grown at 850 °C in H_2+N_2 atmosphere. Particularly, the AlGaN barriers were grown using the ALD technique. The ALD process involves alternate control of mass flow of TMAl and TMGa gases during the growth of AlGaN barrier to form six pairs of AlN/GaN SLs. Figure 1 shows the growth procedure of the AlGaN barrier and GaN well layer. The TMAl and TMGa flow times of AlN and GaN layers were 6.8 and 19.8 s, respectively, under a continuous flow of the NH_3 gas at 850 °C. The growth rate of the ALD grown AlGaN barrier measured by an *in situ* Filmetrics optical monitoring system was about 0.14 μm/h. After the AlGaN barrier was grown, only TMGa was introduced into the reactor for 34.8 s to grow the GaN well.

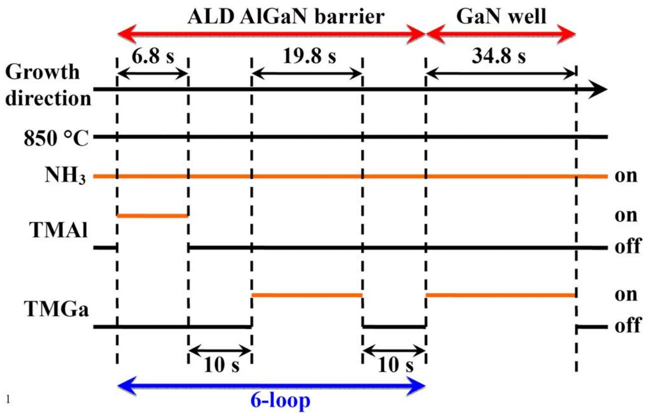

FIG. 1. (Color online) Growth procedure of AlGaN barrier and GaN well layers.

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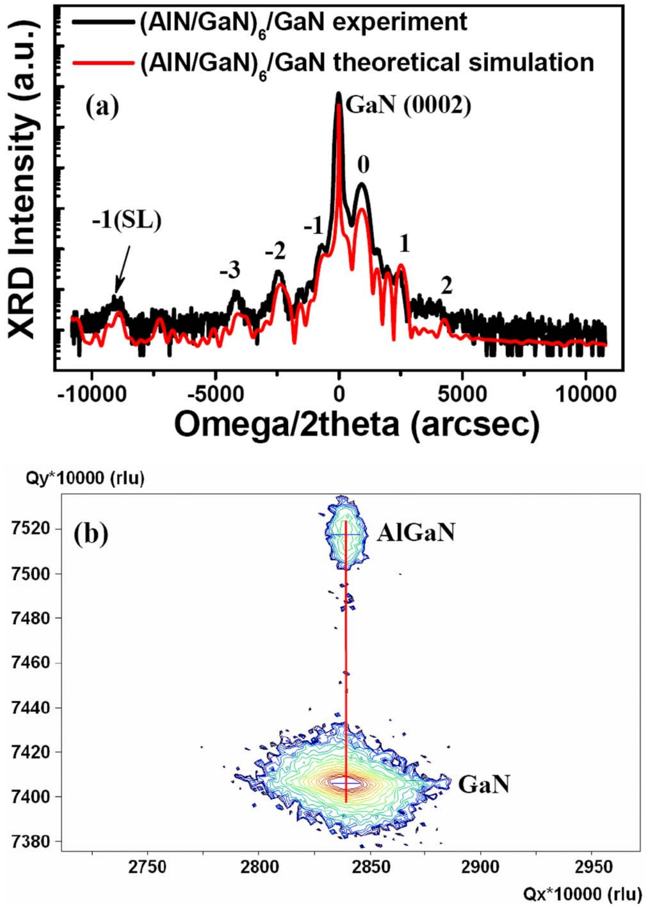

FIG. 2. (Color online) (a) HRXRD pattern at (0002) plane for the AlGaN/GaN MQWs sample, (b) RSM of the sample obtained from (1 0 $\bar{1}$ 5) diffraction.

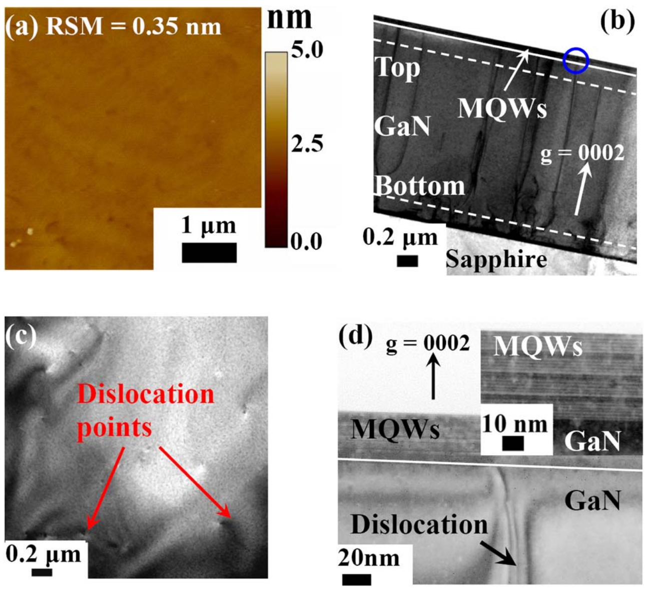

FIG. 3. (Color online) (a) Surface morphology of the grown AlGaN/GaN MQWs sample scanned by AFM; (b) cross-sectional TEM image and (c) plane-view TEM image of the AlGaN/GaN MQWs sample; (d) enlarged cross-sectional TEM images of the sample.

The surface morphology of the as-grown sample was observed by atomic force microscope (AFM) with a scanning area of 5×5 μm^2. Crystalline quality was evaluated by high resolution x-ray diffraction (HRXRD) and reciprocal space mapping (RSM), and Cu $K\alpha$ 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 between satellite peaks in the (0002) $\omega/2\theta$-scan. The optical properties were investigated by PL measurements. PL spectra were excited with a frequency tripled Ti:sapphire laser at wavelength of 266 nm and the laser output power was 20 mW. The laser pulse width was 200 fs and the repetition rate was 76 MHz. The luminescence spectrum was measured by a 0.5 m monochromator and detected by a photomultiplier tube. The CL measurements were carried out at 300 K by using a mono-CL system installed on a field emission scanning electron microscope with beam energies of 5–20 keV. The threading dislocations and the sharpness of the AlGaN/GaN interfaces were studied by transmission electron microscope (TEM). The dislocation density of the sample surface with an area of 20×26 μm^2 was analyzed after the 5 min etching in the KOH solution with 0.005*M* at 80 °C.

Figures 2(a) and 2(b) show the HRXRD $\omega/2\theta$ diffraction pattern and the RSM of the as-grown AlGaN/GaN MQWs sample. In Fig. 2(a), the HRXRD diffraction pattern shows two periodical structures: one can be attributed to the AlGaN/GaN MQWs; another can be attributed to the AlN/GaN SLs in the AlGaN barriers. The third order satellite peak of the diffraction pattern for AlGaN/GaN MQWs can be clearly observed, suggesting the high crystalline quality of AlGaN/GaN MQWs and AlN/GaN SLs. The thickness of AlN and GaN in the barrier and the GaN wells can be fitted to be about 0.42, 0.77, and 2.9 nm, respectively. The average Al content of AlGaN barrier is also estimated to be about 0.29. From the RSM data of AlGaN/GaN MQWs obtained from (1 0 $\bar{1}$ 5) diffraction shown in Fig. 2(b), the spread of RSM intensity for the AlGaN/GaN MQWs was relatively narrow indicating that AlGaN epilayers exhibited relatively small distribution of crystal orientation.[10] In addition, the reciprocal lattice points of AlGaN and GaN were lined up at the same Q_x position (red solid line) indicating the AlGaN and GaN had same lattice constant. According the earlier report,[11] the degree of lattice relaxations can be estimated from the equation of $\varepsilon_{xx}=q_x^{\mathrm{GaN}}/q_x^{\mathrm{MQWs}}-1$, where the q_x^{GaN} and q_x^{MQWs} are the x position of GaN layer and AlGaN layer, respectively. We obtained an estimated degree of lattice relaxation to be only 3.9×10^{-6}, indicating that the AlGaN epilayer is fully strained and pseudomorphic to the underlying GaN layer.

As shown in Fig. 3(a), the surface morphology of the top layer was observed by AFM and no cracks were found. A very small rms value of the surface roughness of 0.35 nm was achieved. To carefully investigate the threading dislocation within our sample, both cross-sectional and plane-view TEM images were taken. Figure 3(b) shows the cross-sectional TEM image of the sample with the white dash lines indicating the top and bottom GaN epilayer regions. It is clear that few dislocations are observable and only one dislocation passes through the GaN epilayer into AlGaN/GaN MQWs. The dislocation density (DD) at the bottom GaN layer is about 3.5×10^{8} cm^{-2} and slightly reduces to 1.4×10^{8} cm^{-2} at the top GaN layer. However, the DD in the AlGaN/GaN MQW region is only 2.5×10^{7} cm^{-2}. Figure 3(c) shows the plane-view bright-field TEM image from the top surface. The DD was estimated to be about 3.2

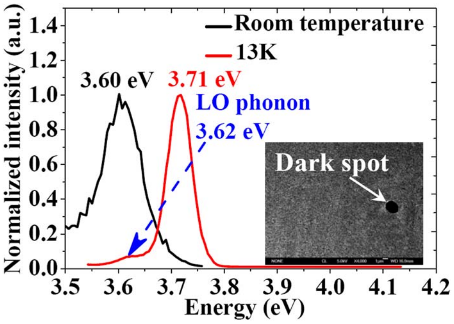

FIG. 4. (Color online) The 13 K and room temperature PL spectra of the AlGaN/GaN MQWs sample. Inset shows CL image taken at $E=3.71$ eV.

$\times 10^7$ cm^{-2}. Meanwhile, we also estimate the DD in this AlGaN/GaN MQWs sample by evaluating the etch-pit density (EPD) of the KOH etched sample. We obtain an estimated EPD value of about 3.3×10^7 cm^{-2}, which is similar to the above estimated plain-view DD value. These estimated DD values of our sample are nearly two orders of magnitude lower than that of AlGaN films, which were not grown by ALD, reported recently.[12] From the enlarged TEM image shown in the inset of Fig. 3(d), it can be clearly observed that the QWs and SLs exhibited sharp interfaces with good periodicity, showing that the high quality SLs and MQWs were formed by the ALD technique. The image also shows that the AlGaN barrier consisted of six pairs AlN/GaN SLs with AlN thickness of 0.43 nm and GaN thickness of 0.77 nm, respectively, forming a AlGaN barrier with thickness of 7.2 nm, and the GaN well had a thickness of 3 nm, which are in good agreement with the result estimated from HRXRD data.

Interestingly, a bending of threading dislocations at the boundary of MQWs without extending into the top surface was commonly observed in this sample, as shown in Fig. 3(d). Previously it was reported[13] that the strain in the epilayer could exert a net force on the dislocation to be bended or terminated at the strained epilayer edge without threading through the epilayer to the top surface. Since our RSM result demonstrated that the AlGaN epilayer is fully strained, it suggested that the large strain in the ALD grown AlGaN barrier with AlN/GaN SLs could effectively bend and suppress the threading dislocations, thus reducing the defects in MQW and improving the surface morphology of the sample.

Figure 4 shows the PL spectra of the as-grown sample. The emission peak energy at 3.60 and 3.71 eV was observed at room temperature and 13 K, respectively. The full width at half maximum of PL spectra is about 80 meV at room temperature and reduces to only 47 meV at 13 K, which are smaller than the previous report by a factor of 2–3,[14] indicating that the crystal quality of AlGaN/GaN MQWs has been improved by using ALD AlGaN barrier. Our PL data analysis confirmed that the two dominant emission peak energies of 3.60 and 3.71 eV at room temperature and 13 K, respectively, was emitted from GaN well. In addition, the emission peak energy at 3.62 eV can be clearly observed at 13 K. According to previous report, the emission peak energy of 3.62 eV could be attributed to the LO phonon.[15] Additionally, the inset of Fig. 4 shows the CL image of the sample which has near uniform brightness with few dark spots. It was well known that the dark spots in CL image were related to nonradiative centers in the defects of epilayers. Therefore the CL image data again suggest our sample has relatively low DD and superior crystalline quality.

In summary, we have grown low dislocation and high crystalline quality AlGaN/GaN MQWs on sapphire substrate by using the ALD grown AlGaN barrier consisted of AlN/GaN SLs. The AFM data show smooth surface morphology with a small surface roughness RMS value of about 0.35 nm and no surface cracks. The TEM and HRXRD measurements show that the grown sample has sharp interfaces between SL layers and QWs with good periodicity. The sample has near uniform CL image intensity at room temperature and narrow PL emission peak. The sample has a low DD of about 3.3×10^7 cm^{-2}. These results indicate that the AlGaN/GaN MQWs grown by the ALD technique is a viable method for growth of a device-quality AlGaN/GaN MQWs structure for various optical devices.

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[1] N. Iizuka, K. Kaneko, N. Suzuki, T. Asano, S. Noda, and O. Wada, Appl. Phys. Lett. **77**, 648 (2000).
[2] Ü. Özgür, H. O. Everitt, L. He, and H. Morkoç, Appl. Phys. Lett. **82**, 4080 (2003).
[3] T. J. Schmidt, X. H. Yang, W. Shan, J. J. Song, A. Salvador, W. Kim, Ö. Aktas, A. Botchkarev, and H. Morkoç, Appl. Phys. Lett. **68**, 1820 (1996).
[4] J. Han, M. H. Crawford, R. J. Shul, J. J. Figiel, M. Banas, L. Zhang, Y. K. Song, H. Zhou, and A. V. Nurmikko, Appl. Phys. Lett. **73**, 1688 (1998).
[5] J. M. Redwing, D. A. S. Loeber, N. G. Anderson, M. A. Tischler, and J. S. Flynn, Appl. Phys. Lett. **69**, 1 (1996).
[6] T. Takano, Y. Narita, A. Horiuchi, and H. Kawanishi, Appl. Phys. Lett. **84**, 3567 (2004).
[7] V. Adivarahan, W. H. Sun, A. Chitnis, M. Shatalov, S. Wu, H. P. Maruska, and M. A. Khan, Appl. Phys. Lett. **85**, 2175 (2004).
[8] Y. Kawakami, X. Q. Shen, G. Piao, M. Shimizu, H. Nakanishi, and H. Okumura, J. Cryst. Growth **300**, 168 (2007).
[9] Y. Kawakami, A. Nakajima, X. Q. Shen, G. Piao, M. Shimizu, and H. Okumura, Appl. Phys. Lett. **90**, 242112 (2007).
[10] Y. S. Park, C. M. Park, S. J. Lee, H. Im, T. W. Kang, J.-E. Oh, C. S. Kim, and S. K. Noh, Semicond. Sci. Technol. **20**, 775 (2005).
[11] G. S. Huang, T. C. Lu, H. H. Yao, H. C. Kuo, S. C. Wang, C.-W. Lin, and L. Chang, Appl. Phys. Lett. **88**, 061904 (2006).
[12] D. M. Follstaedt, A. A. Allerman, S. R. Lee, J. R. Michael, K. H. A. Bogart, M. H. Crawford, and N. A. Missert, J. Cryst. Growth **310**, 766 (2008).
[13] J. Knall, L. T. Romano, D. K. Biegelsen, R. D. Bringans, H. C. Chui, J. S. Harris, Jr., D. W. Treat, and D. P. Bour, J. Appl. Phys. **76**, 2697 (1994).
[14] H. Hirayama, J. Appl. Phys. **97**, 091101 (2005).
[15] M. Smith, J. Y. Lin, H. X. Jiang, A. Salvador, A. Botchkarev, W. Kim, and H. Morkoc, Appl. Phys. Lett. **69**, 2453 (1996).