Fabrication and Characteristics of GaN/AlGaN Multilayer Structure for Terahertz Quantum-Cascade Laser

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ABSTRACT

The GaN/AlGaN multilayer structure for the active regions of terahertz quantum cascade lasers (QCLs) was grown by metal organic chemical vapor deposition (MOCVD). The surface morphology of the grown sample showed good surface quality with an average roughness of less than 1 nm. The x-ray diffraction pattern and transmission electron microscopy images showed that the well-controlled quantum cascade GaN/AlGaN layers were grown. The Fourier transform infrared spectrometer measurement showed a distinct A_1 (LO) phonon frequency at 822 cm^{-1} that is red-shifted with respect to the single Al$_{0.2}$Ga$_{0.8}$N layer due to the good periodicity of the grown quantum cascade GaN/Al$_{0.2}$Ga$_{0.8}$N structure. MOCVD growth should be a viable technique for fabrication of AlGaN/GaN quantum cascade laser and phonon frequency shift should be a key indicator for the good periodicity of the grown QCL structure.

Keywords: Quantum cascade laser, MOCVD, AlGaN, TEM, XRD

1. INTRODUCTION

Recently considerable effort has been expanded to develop a solid-state coherent source in the THz range. Several semiconductor intersubband transitions have been proposed as potential candidates for THz lasers [1-3] and were recently demonstrated in the GaAs/AlGaAs material system using a quantum cascade laser (QCL) scheme [4-11]. In the GaAs-based QCLs, the energy separation between the lower laser state and the ground state is just above the LO-phonon energy (~36 meV), which is comparable to room temperature k_BT (~26 meV). Obviously, a material system with larger LO-phonon energy will be desirable for the high-temperature operation of THz quantum cascade lasers. G. Sun et al.[12] proposed to use a GaN-based system with large LO-phonon energy (~90 meV) for THz QCLs. The advantages are threefold. First, the large LO-phonon energy of ~ 90 meV in a GaN-based system can reduce the thermal population of the lower laser state. Second, ultrafast LO-phonon scattering in GaN/AlGaN quantum wells can be used for the rapid depopulation of the lower laser state [12, 13]. Third, the large LO-phonon energy can also increase the lifetime of the upper laser state by reducing the relaxation of electrons with higher in-plane kinetic energy via emission of a LO-phonon. G. Sun et al. [12] analysis of their structure showed that the laser can have a relatively low threshold current density of 832 A/cm^2 with a threshold optical gain of 50/cm at room temperature and predicted the laser has a characteristic temperature as high as 136K.

Although fabrication of AlGaN/GaN quantum cascade laser structures has been reported, these samples were grown by molecular beam epitaxy (MBE) and hot wall epitaxy (HWE) [14, 15]. Metal organic chemical vapor deposition (MOCVD) should offer several advantages over MBE and HWE for QCL fabrication, including higher growth rates, easier reactor maintenance, and mass production. In this report, we present the results of our growth of AlGaN/GaN QCL active layer structure using MOCVD system and the investigation and analysis of the grown structure to establish the compositional contents and thickness of the grown active layer structures which are very important parameters for the performance of GaN QCLs. We used a MOCVD system to grow GaN/AlGaN active region multilayer structures for THz QCL as designed by G. Sun et al. [12]. Atomic force microscopy (AFM), X-ray diffraction (XRD), transmission electron microscopy (TEM) and Fourier transform infrared (FTIR) spectrometer were used to investigate and characterize the grown samples. The grown samples showed smooth surface morphology with abrupt layer interfaces.

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Relatively good control of Al composition and layer thickness was demonstrated. The slight frequency shift of the A_1(LO) phonon in the multilayer structure was observed.

2. EXPERIMENTS

All the samples in this work were grown in a low-pressure vertical (EMCORE D75) reactor with a high speed rotation of the susceptor. The reagents were pure ammonia, trimethylgallium (TMGa) and trimethylaluminum (TMAl). Hydrogen and nitrogen purified by purifier were used as the carrier gases. The rotating speed was 900 rpm. C-plane sapphire epi-ready substrates were used for all the growth.

The group III and V precursors were separated in two manifolds and mixed before they entered the reactor. Prior to material growth, the sapphire substrate was annealed to remove any residual impurities on the surface in H_2 ambient at 1100 ^{o}C for 5 min. For all samples, a normal 30-nm-thick GaN nucleation layer was deposited at 500 ^{o}C. Then the temperature was raised up to 1100 ^{o}C for growth of a 2-μm-thick GaN buffer layer. Before the structures were grown, the ambient gas was changed into nitrogen with hydrogen and the ratio of two carrier gases keeps constant. The schematic structure of the active region of quantum cascade laser is shown in Fig.1. The full structure consisted of 20 periods of quantum wells with each period consisting of 3 GaN QWs and 3 $Al_{0.2}Ga_{0.8}N$ barriers with layer thickness (nm): 3/**4**/3/**2.5**/2/**2.5** (wells in bold and barriers in plain). The surface morphology and epitaxial thickness of multi-layer were measured by atomic force microscopy (AFM) and transmission electron microscopy (TEM). The X-ray diffraction pattern of the active regions was measured and simulated by using a Bede Scientific D1 double crystal X-ray diffraction. The infrared reflectivity spectra were collected at room temperature by Bomen Fourier transform infrared spectrometer. We used nonpolarized light with an incident angle of 75^{o} (Brewster's angle was about 68^{o}) and a Fourier transform spectrometer, equipped with KBr beam splitter and a mercurcy-cadmium-tellride detector cooled down to 77K.

3. RESULTS AND DISCUSSION

Figure 2 shows an AFM image of the grown sample. The average roughness measured by AFM was less than 1 nm over a 5×5 μm^2 surface area, which was comparable to that of high quality GaN epilayer.

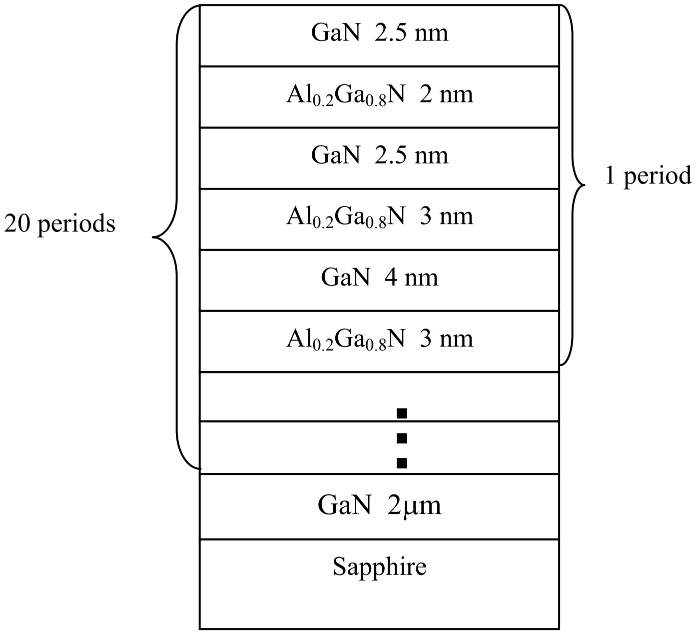
Figure 3 shows a cross-sectional TEM image of the 20-period QC structure and each period consisting of 3 GaN QWs and 3 $Al_{0.2}Ga_{0.8}N$ barriers. In Figure 3 (a), a contrast made by the periodically aligned 20 sets of the MQWs can be seen. No dislocations running across the sample were observable in this figure. In Figure 3 (b), the lighter parts and darker parts correspond to the AlGaN barriers and the GaN wells, respectively. It can be seen that the interfaces are very sharp. The actual layer thickness of the GaN QWs and $Al_{0.2}Ga_{0.8}N$ barriers can be estimated from the TEM pictures. The thickness difference between the grown sample and the designed structure was less than 0.5 nm.

The crystalline quality of the samples was evaluated by (0002) symmetric high-resolution x-ray diffraction (HRXRD) with a Cu K_{a1} radiation. The average thicknesses of the AlGaN barriers and the GaN wells were determined by the angular distance using satellite peaks of ω/2θ-scan diffraction patterns. The HRXRD pattern of the ω/2θ-scan (0002) reflections for QC structure is shown in Fig. 4. The top and bottom lines show the experimental and simulated results, respectively. The satellite peaks due to the QC structure can be clearly seen up to the 4th order peaks indicates that smooth and abrupt interfaces with good periodicity of the QC structure for the 20 cascading periods. Table 1 listed the simulated thicknesses and Al compositions of epilayers in one period. The good agreement between the experimental curve and the simulated data confirmed the successful fabrication of the QC structure.

Figure 5 shows measured FTIR spectra for a GaN film, an $Al_{0.2}Ga_{0.8}N$ with a GaN buffer and a QCL structure with a GaN buffer deposited on sapphire substrates. Two dips labeled as β, γ with frequencies of 758 cm^{-1} and 890 cm^{-1}, respectively, are originated from the GaN and sapphire [16]. The dips labeled with dashed line were assigned as an AlN like A_1 (LO) phonon. For $Al_{0.2}Ga_{0.8}N$ with GaN buffer, the phonon frequency is about 839 cm^{-1}, while for grown QCL structure, the frequency of the A_1 (LO) phonon is about 822 cm^{-1} which is shifted by about 17 cm^{-1} as shown in Figure 5. Since there are sharp interface between barrier and well layers of the GaN/AlGaN structure in the grown QCL active layer that could cause increase in the real space periodicity resulting in reduction of Brillouin zone, the frequency shift of A_1 (LO) phonon could be due to the well known zone-folding effect [17]. The frequency shift of A_1 phonon mode should be a key indicator for good periodicity of grown quantum cascade laser structure.

4. CONCLUSIONS

In summary, we have grown high crystalline quality active regions of $Al_{0.2}Ga_{0.8}N$/GaN QCL by low-pressure MOCVD. The morphologies of full active regions were quite smooth. The compositional data of $Al_xGa_{1-x}N$ were determined and the active layer structure of QCL was successfully grown. The grown QCL structure showed smooth surface morphology with well-controlled Al compositions and thicknesses. The FTIR measurement of the QCL GaN/AlGaN structure showed clear A_1 (LO) phonon mode with a frequency around 822 cm^{-1} which is red-shifted with respect to the single layer $Al_{0.2}Ga_{0.8}N$ layer due to the good periodicity of the grown active GaN/$Al_{0.2}Ga_{0.8}N$ structure.



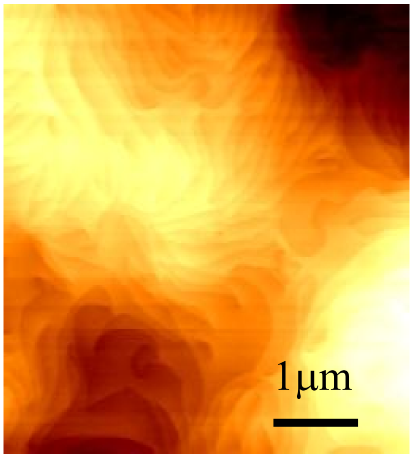


Figure 1 Schematic diagram of AlGaN QCL active layer structure

Figure 2 AFM top-view image of grown AlGaN/GaN QCL structure

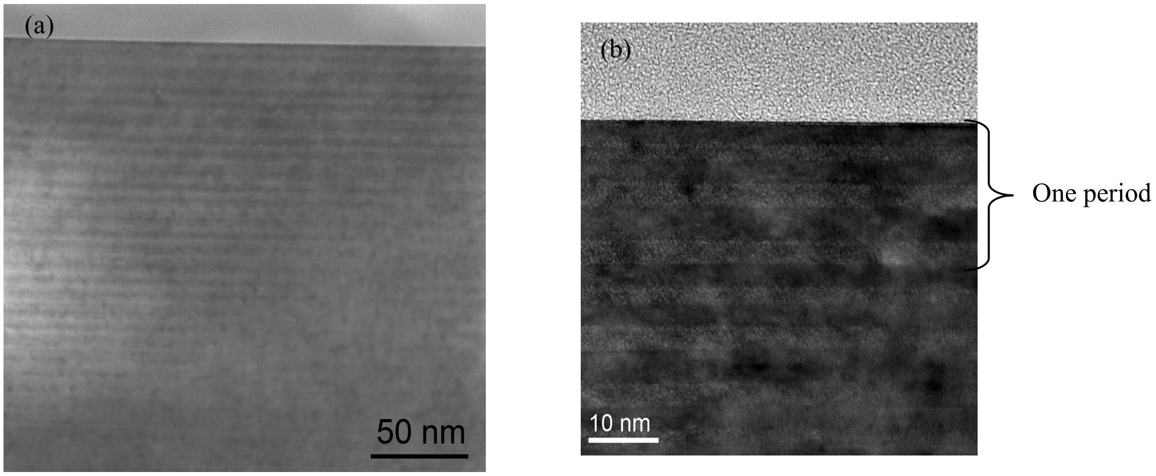


Figure 3 (a) cross-sectional TEM images of 20 periods MQW sample. (b) two periods with each period consisting of 3 GaN QWs and 3 AlGaN barriers with layer thickness (nm): 3/**4**/3/**2.5**/2/**2.5** (wells in bold and barrier in plain).

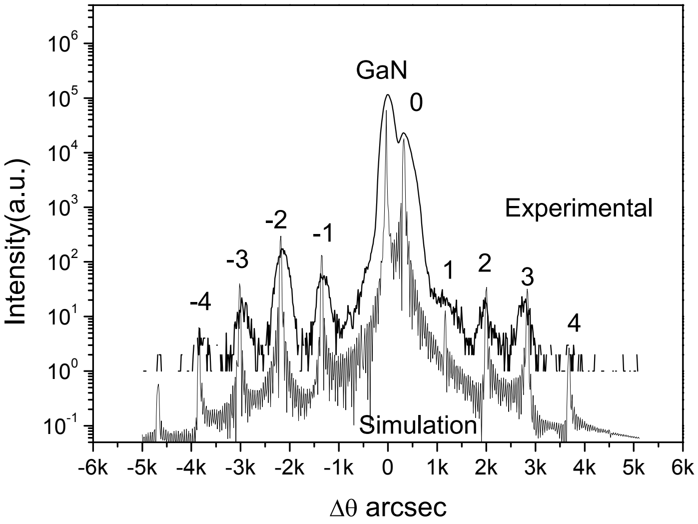


Figure 4 (0 0 0 2) ω-2θ x-ray diffraction pattern and simulated result of AlGaN/GaN quantum cascade laser active region structure.

Table 1. Simulation results of x-ray diffraction pattern

layer	Thickness (nm)	Material	composition
1	2.9	GaN	
2	2.4	$Al_xGa_{1-x}N$	0.20
3	2.9	GaN	
4	3.6	$Al_xGa_{1-x}N$	0.20
5	4.5	GaN	
6	3.6	$Al_xGa_{1-x}N$	0.20

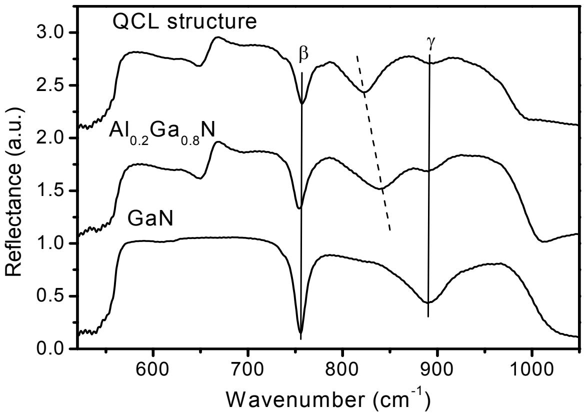


Figure 5. Room temperature FTIR of QCL structure, $Al_{0.2}Ga_{0.8}N$ epilayer and GaN bulk

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