HRTEM investigation of high-reflectance AlN/GaN distributed Bragg-reflectors by inserting AlN/GaN superlattice

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\textbf{Abstract}
A high-quality AlN/GaN distributed Bragg-reflectors (DBR) was successfully grown on sapphire substrate by low-pressure metal-organic chemical vapor deposition using ultra-thin AlN/GaN superlattice insertion layers (SLILs). The reflectivity of AlN/GaN DBR with ultra-thin AlN/GaN SLIL was measured and achieved blue peak reflectivity of 99.4\% at 462 nm. The effect of ultra-thin AlN/GaN superlattice insertion layer was examined in detail by transmission electron microscopy, and indicated that the crack of AlN/GaN DBR can be suppress by inserting AlN/GaN SLIL. For electronic properties, the turn on voltage is about 4.1 V and CW laser action of vertical-cavity surface-emitting laser (VCSEL) was achieved at a threshold injection current of 1.4 mA at 77 K, with an emission wavelength of 462 nm.

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\section{Introduction}
The heteroepitaxy of monolithic distributed Bragg-reflectors (DBR) of III-nitride material system is of great interest because of its potential applications, including in resonant-cavity light-emitting diodes (RCLEDs) and vertical-cavity surface-emitting lasers (VCSELs)\textsuperscript{[1,2]}. Nevertheless, it is relatively difficult to grow high-quality III-nitride-based DBR on the sapphire substrate due to the lattice mismatch and misfit in thermal expansion coefficients between these two material systems. In addition, some reports indicated that the optical and electrical properties of DBR were very sensitive to the threading dislocation density (TDD) in DBR suggesting high crystalline quality DBR could dramatically improve the performance. So far, a few of studies have attempted to grow III-nitride-based DBR with ultra-flat interfaces and low threading dislocation density therein on a variety of substrates. Recently, we have grown high-quality AlN/GaN DBR on sapphire by simply using an ultra-thin superlattices insertion layer (SLIL), in which AlN and GaN (AlN/GaN) were deposited as the low index layer in AlN/GaN DBR\textsuperscript{[3]}. Simultaneously, we have conducted analysis and recognize the improvement in the material quality of AlN/GaN DBR with ultra-thin AlN/GaN SLIL. For the verification of dislocation density in AlN/GaN DBR, apart from chemical etching, transmission electron microscopy (TEM) is well-known to be another effective approach to determine the density of dislocations. In particular, an image obtained directly by TEM cannot only be used to evaluate accurately the dislocation density but also to elucidate the kinetics of the defects. Therefore, to further recognize our AlN/GaN DBR with AlN/GaN superlattice insertion layer, in this article, we examine the effect of ultra-thin AlN/GaN superlattice insertion layer on the epitaxial growth of AlN/GaN DBR by analysis of the structural, electronic and optical properties.

\section{Experiment}
The epitaxial growth of AlN/GaN on sapphire substrate with ultra-thin AlN/GaN SLIL was performed using a low-pressure EMCORE D75 metal-organic chemical vapor deposition (MOCVD) system with a vertically cold-wall chamber. TMGa, TMAl and NH\textsubscript{3} were the sources of Ga, Al and N, respectively. N\textsubscript{2} and H\textsubscript{2} were the carrier gases. The temperature was monitored using a thermocouple near the graphite susceptor. The substrates used in the experiment were cut from (0\,0\,0\,1)-oriented sapphire wafers. Before loading, the substrate was cleaned by acetone and then rinse by deionized water to obtain a stable and clean surface. After
loading, the sapphire substrate was thermally etched at 1000 °C in an H2 environment. Following the thermal etching, a nucleation layer of GaN was first deposited, and then the top un-doped epilayer was grown. Finally, the AlN/GaN DBR with ultra-thin AlN/GaN SLIL was grown atop the un-doped GaN epilayer. The further growth details are shown in Table 1. In addition, the growth of specimens without AlN/GaN SLIL was also conducted for comparison. The epitaxial structures of two samples are shown in Fig. 1 [3]. Finally, the VCSEL devices were fabricated and the fabrication procedure of VCSEL devices was reported elsewhere [4].

The distribution and threading behaviors of dislocations in AlN/GaN DBR were then studied by transmission electron microscopy. Moreover, the interfacial microstructures of the epilayer were observed by high-resolution TEM. The reflectivity spectra of the GaN/AlN DBRs were measured by the n and k ultraviolet–visible spectrometer with normal incidence at room temperature. Finally, the current–voltage (I–V) and light–current (L–I) characteristics of VCSEL devices with high-quality AlN/GaN DBR were measured at 77 K.

### 3. Results and discussion

Fig. 2 presents cross-sectional TEM images of AlN/GaN DBR grown on sapphire substrate (a) with and (b) without AlN/GaN SLIL. Comparing with the specimen without AlN/GaN SLIL, evidently the interface of AlN/GaN DBR becomes sharper when AlN/GaN SLIL was used. In addition, from Fig. 2, the threading dislocation density in GaN epilayer for these two samples can be estimated. Here, the two blue dash lines indicated the top and bottom region of GaN epilayer, respectively. So the TDD at the bottom and top of GaN layer is about \(9.0 \times 10^8\) and \(3.9 \times 10^9\) cm\(^{-2}\) for sample with AlN/GaN SLIL; however, the TDD at the bottom and top of GaN layer is about \(1.2 \times 10^9\) and \(8.6 \times 10^9\) cm\(^{-2}\) for sample without AlN/GaN SLIL. On the other hand, to confirm the crystalline quality of DBR and observe more clearly the dislocation reduction behavior in the epilayer, both the magnifications of regions I in Fig. 2(a) and (b) were taken and presented in Fig. 2(c) and (d), respectively. As can be seen in Fig. 2(d), there were many V-shape defects accompany the crack line in the DBR region. However, for AlN/GaN DBR grown by inserting AlN/GaN SLIL, although there were many V-shape defects in BDR region, no crack line can be found throughout the observed area. Hence, the results of above-mentioned indicate that the crystalline quality of the AlN/GaN DBR grown by the proposed method was much better than that grown by the conventional method without AlN/GaN SLIL.

Fig. 3 shows the high-revolution TEM image of AlN/GaN SLIL. As can be seen, the SLIL shows smooth and sharp interface between AlN and GaN SLs and interface between AlN and GaN indicated by the white dash line in the Fig. 3. In addition, the elemental composition of AlN and GaN in SLIL were evaluated by the in-situ energy-dispersive spectroscopy (EDS), which was installed on the TEM system. Therefore, we can found that the Al composition of region A and region B in Fig. 3 are about 50.08% and 12.76%, respectively. Here a set of GaN/AlN SLIL can be seen as a digital alloy of an \(\text{Al}_x\text{Ga}_{1-x}\)N layer \(x\sim0.45\) for a low reflective index quarter-wave layer.

Fig. 4 shows reflectivity spectra of AlN/GaN DBR grown on sapphire with and without AlN/GaN SLIL. It can be found that the reflectivity spectral of the specimen without AlN/GaN SLIL exhibited non-flat top stop band, and the reflectivity at 464 nm and the stop band width are about 95.3% and 26 nm, respectively. However, for the specimen with AlN/GaN SLIL, the reflectivity spectral exhibited flat top stop bands, and the reflectivity at 462 nm and the stop band width are about 99.4% and 29 nm, respectively. Hence, this data indicated high crystal quality of the specimens and is probably connected to that of crack-free. In our experiment, we found that the TDD of GaN epilayer for sample with SLIL is relatively close to the sample without SLIL. Additionally, many V defects are found in the DBR region for sample with and without SLIL, as shown in Fig. 2(c) and (d), respectively. According to the report by Cho’s et al. [5], V defects mainly arose from the stacking mismatch boundaries and threading dislocation. In other words, there are many threading dislocations which can be radiating vertically from the interface between GaN and sapphire into the AlN/GaN DBR region in spite of the sample with AlN/GaN SLIL. As a result, the TDD of underlayer GaN in these two samples shall not result in the difference of reflectance. However, when the AlN/GaN SLIL was used, we clearly found that the crack of AlN/GaN DBR was reduced.
drastically compared with the sample without AlN/GaN SLIL, as shown in the blue circle of Fig. 2(c) and (d). In addition, our previous report confirmed that the sample with SLIL exhibited smaller in-plane strain, and no crack can be observed on the surface of sample with SLIL as AlN/GaN SLIL was used [3]. Besides, it was well known that the large lattice mismatch simultaneously with the high thermal coefficient incompatibility between AlN and GaN can result in a relatively small critical thickness, leading to the formation of crack. In other words, the AlN/GaN SLIL can be seen as a digital alloy of an Al$_x$Ga$_{1-x}$N layer $x \approx 0.45$. That is to say, the AlN/GaN SLIL can reduce the lattice mismatch between GaN and AlN, and then the critical thickness of epilayer was increased whereby the crack in AlN/GaN DBR was suppressed and reduced.
Therefore, based on our results, the suppression of crack formation could be responsible for the high-reflectance AlN/GaN DBR with AlN/GaN SLIL.

Finally, the GaN-based blue VCSEL devices fabricated based on the material structure of AlN/GaN bottom DBR grown by inserting AlN/GaN SLIL. Fig. 5 gives the light output power versus CW injection current and current–voltage characteristics of the VCSEL sample at 77 K. The turn on voltage is about 4.1 V, indicating the ohmic contact with low resistance was performed since good electrical contact of the ITO transparent contact layer and the intracavity current injection scheme. A dominant single laser emission line at 462 nm appears above the threshold current. Additionally, the threshold current, \( I_{th} \), is about 1.4 mA and then was linearly increased with the injection current beyond the threshold. The threshold current density is estimated to be about 1.8 kA/cm² for a current injection aperture of 10 \( \mu \)m in diameter.

4. Conclusions

In summary, high reflectivity and crack-free AlN/GaN DBR have been grown successfully on sapphire by low-pressure MOCVD using AlN/GaN SLIL. The effect of ultra-thin AlN/GaN superlattice insertion layer was examined in detail by transmission electron microscopy, and indicated that the suppression of crack formation and reduction of threading dislocation density of AlN/GaN DBR were promoted by inserting AlN/GaN SLIL. Additionally, the reflectivity of AlN/GaN DBR with ultra-thin AlN/GaN SLIL was measured and achieved blue peak reflectivity of 99.42% at 462 nm. For VCSEL devices, the turn on voltage is about 4.1 V and CW laser action of VCSEL was achieved at a threshold injection current of 1.4 mA at 77 K, with a lasing wavelength of about 462 nm. Further optimization of AlN/GaN DBR with ultra-thin AlN/GaN SLIL grown by low-pressure MOCVD makes it possible to grow high reflectivity and crack-free DBR on sapphire to achieve CW lasing of GaN-based VCSEL devices at room temperature.

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References