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# Growth of AlGa<sub>N</sub>/Ga<sub>N</sub> Quantum Wire Structures by Radio-Frequency-Radical-Assisted Selective Molecular Beam Epitaxy on Prepatterned Substrates

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The feasibility of the selective molecular beam epitaxy (MBE) growth of AlGa<sub>N</sub>/Ga<sub>N</sub> quantum wire (QWR) structures on prepatterned substrates is investigated. The detailed studies on growth features have revealed that size-reducing selective growth is possible on mesa patterns having the <11-20>-orientation, but not on those having the <1-100>-orientations. The behavior reflects complex growth kinetics on high-index crystalline facets. The lateral wire width of QWR structures formed selectively on a top mesa can be controlled by adjusting the growth thickness and the initial size of mesa patterns. From cathodoluminescence (CL) measurements, emission from the embedded AlGa<sub>N</sub>/Ga<sub>N</sub> QWR structure has been clearly identified.

**KEYWORDS:** selective growth, molecular beam epitaxy (MBE), patterned substrates, Ga<sub>N</sub>, AlGa<sub>N</sub>

## 1. Introduction

Recently, the AlGa<sub>N</sub>/Ga<sub>N</sub> system has been opened up and has led to the establishment of blue/UV photonic and high-power electronic device application areas. This material system, however, is potentially suitable for not only these areas but also for high-operating-temperature quantum devices such as quantum-wire transistors (QWR-Trs) and single-electron transistors (SETs). This is due to wide energy gaps with a large  $\Delta E_c$  and to the availability of high-density two dimensional electron gases (2DEGs) even under non-doped conditions which avoids the doping fluctuation problem commonly observed in nanometer-scale devices. Here, a key question is how to form the AlGa<sub>N</sub>/Ga<sub>N</sub>-based nanostructures in a size- and position-controlled manner. Recent studies on the fabrication of Ga<sub>N</sub>-based quantum structures have been mostly focused on the self-assembled formation of quantum dots (QDs) using the Stranski-Krastanov mode [1,2] or anti-surfactant methods [3,4]. In these methods, control of size and position seems to be very difficult. On the other hand, we have shown that selective molecular beam epitaxy (MBE) growth on a prepatterned substrate is a very powerful approach for the formation of high-density quantum nanostructures for GaAs- and InP-based materials [5-7] with tight control of both position and size.

The purpose of this study is to investigate the basic behavior and feasibility of the selective MBE growth of AlGa<sub>N</sub>/Ga<sub>N</sub> quantum wire (QWR) structures on prepatterned substrates for the first time. For this purpose, Ga<sub>N</sub> and AlGa<sub>N</sub> layers were grown by rf-radical-assisted MBE growth. Through growth on planar Ga<sub>N</sub> (0001) substrates, the basic growth conditions were optimized using in situ reflection high-energy electron diffraction (RHEED) and X-ray diffraction (XRD) analyses. Then, selective growth experiments were

carried out on patterned substrates. The structural and optical properties of the selectively grown GaN wires were characterized by scanning electron microscopy (SEM) and cathodoluminescence (CL) techniques. CL measurements were performed at 7 K using a cryostat.

A detailed study on the cross-sectional features of the grown structure has indicated that size-reducing selective growth is possible on mesa patterns having the  $\langle 11\text{-}20 \rangle$ -orientation. The lateral wire width of QWR structures can be controlled by adjusting the growth thickness of the AlGaIn and the initial size of mesa patterns. From cathodoluminescence (CL) measurements, the emission from the embedded AlGaIn/GaN QWR structures has been clearly identified.

## 2. MBE Growth of GaN and AlGaIn on Planar Substrates

### 2.1 Growth procedure

In order to optimize the conditions for rf-radical-assisted MBE growth of GaN and AlGaIn, uniform growth experiments were repeated on planar (0001) GaN substrates. Nitrogen was supplied using an rf radical source exited with a microwave power of 350 W. In all experiments, the  $\text{N}_2$  flow rate and growth temperature,  $T_{\text{sub}}$ , were set to be 0.5 sccm and  $800^\circ\text{C}$ , respectively.

Prior to MBE growth, organic cleaning and HF treatment were applied in the atmosphere. Just before the MBE growth, thermal cleaning under  $\text{N}_2$  radical pressure was carried out in the MBE chamber for 5 min. From detailed observations of RHEED patterns, cleaning at a substrate temperature,  $T_{\text{sub}}$ , around  $800^\circ\text{C}$  was found to be most effective in removing the native oxide or other contaminations from the surface.

### 2.2 Results of uniform growth

The measured growth rates of GaN and AlN layers are plotted in **Fig. 1(a)** as functions of the K-cell temperatures of the Ga source,  $T_{\text{Ga}}$ , and Al source,  $T_{\text{Al}}$ . The substrate temperature during growth was  $T_{\text{sub}} = 800^\circ\text{C}$ . The dashed lines in **Fig. 1(a)** show the expected group III limited rates of growth from the Arrhenius relationships for Ga and Al beam supplies from K-cells.

As for the growth of the GaN layer, the growth rate increased with  $T_{\text{Ga}}$  and showed saturation at around  $T_{\text{Ga}} = 900^\circ\text{C}$  with a rate of 100~150 nm/h, as seen in **Fig. 1(a)**. This indicates that the growth became rate-limited by the V-group supply where the supply rate of Ga molecules became larger than that of nitrogen. In situ RHEED observation showed that well-defined bright streak (2x2) patterns, such as those shown in **Fig. 1(b)**, were observed and maintained within the range of  $T_{\text{Ga}} = 900 \sim 1000^\circ\text{C}$ . This result suggests that excess Ga adatoms on the growth surface easily desorb into vacuum, leading to the formation of a smooth surface even under the Ga-rich condition. On the other hand, the growth rate of the AlN layer did not show saturation over a wide range of  $T_{\text{Al}} = 950^\circ\text{C} \sim 1100^\circ\text{C}$ . However, at a high cell temperature  $T_{\text{Al}}$  of  $1100^\circ\text{C}$ , the (2x2) RHEED pattern obtained at lower cell temperatures became dark and spotty, indicating the growth of an Al-rich poor-quality layer. From in situ RHEED observations, it was found that a growth rate within 100 ~ 150 nm/h was suitable for the growth of both GaN and AlN layers.

The growth behavior of AlGaIn layers is summarized in **Fig. 2** where the incorporation rates of group III atoms into the planar (0001) epitaxial layer are plotted as functions of the Al atom supply rate.  $T_{\text{sub}}$  was set to be  $800^\circ\text{C}$ . The Ga supply rate was maintained at a constant value at  $T_{\text{Ga}} = 940^\circ\text{C}$ . Here, the incorporation rates of group III atoms were determined by thickness measurements by cross-sectional SEM observation and Al composition measurements by XRD analyses. As seen in **Fig. 2**, the total amounts of Al and Ga adatoms incorporated into the surface remained constant in spite of the increase in Al supply rate, resulting in a corresponding decrease in the incorporation rate of Ga adatoms. This surprising

result can be explained in terms of the group V supply limited growth in combination with the fact that the incorporation rate of Al adatoms is larger than that of Ga adatoms.

From the XRD analyses, it was found that the Al composition of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  layers could be changed from  $x = 0.24$  to  $0.43$  by changing the Al source temperature,  $T_{\text{Al}}$ . A minimum FWHM value of the XRD peak of 300 s was obtained for both GaN and  $\text{Al}_{0.24}\text{Ga}_{0.76}\text{N}$  layers grown at  $T_{\text{Ga}}=940^\circ\text{C}$  and  $T_{\text{Al}}=980^\circ\text{C}$ , respectively. This value was equal to or even smaller than that of the MOVPE GaN template used in this study.

### 3. Selective Growth of AlGaN/GaN Wire Structures

#### 3.1 Growth method

The sequence of selective growth is schematically shown in **Fig. 3**. As the template for selective growth, straight mesa stripes, as shown in **Fig. 3(a)** were formed by electron-beam (EB) lithography and etching on MOVPE (0001) GaN/ sapphire substrates. Since wet chemical etching is difficult for III-nitride materials, etching was carried out by an electron cyclotron resonance reactive ion beam etching (ECR-RIBE) process developed for nitrides by our group [8]. It uses a methane-based gas mixture of  $\text{CH}_4/\text{H}_2/\text{Ar}/\text{N}_2 = 5/15/3/3$  sccm where the addition of  $\text{N}_2$  is extremely important. ECR plasma was exited at a microwave frequency of 2.45 GHz with a power of 250 W. In order to determine the suitable mesa orientations for the formation of quantum nanostructures,  $\langle 1-100 \rangle$  and  $\langle 11-20 \rangle$  directions were investigated in this study, and the properties of selective growth for these mesa orientations were compared.

Then, selective growth of GaN-based QWR structures was attempted by depositing the materials shown in **Fig. 3(b)** onto the mesa-patterned substrate. First, a GaN buffer layer was grown on the patterned substrates. Then, an  $\text{Al}_{0.24}\text{Ga}_{0.76}\text{N}/\text{GaN}/\text{Al}_{0.24}\text{Ga}_{0.76}\text{N}$  sandwiched layer layers was deposited on the GaN buffer mesa, leading to the formation of embedded GaN wires, as schematically shown in **Fig. 3(c)**.

#### 3.2 Growth selectivity on patterned substrates

Cross-sectional SEM images of the  $\langle 1-100 \rangle$ - and  $\langle 11-20 \rangle$ -oriented mesa patterns fabricated by the ECR-RIBE etching process are shown in **Figs. 4(a), 4(b) and 4(c)**. In this study, the etching depth of 450 nm was used for both orientations by controlling the etching time. After etching, well-defined crystalline facets appeared as sidewalls of the mesa stripe. As shown in **Fig. 4(b)**, the mesa structures defined by two (11-22) side facets were obtained for the stripe in the  $\langle 1-100 \rangle$  direction. On the other hand, the mesa structures defined by two (1-101) facets were obtained in the  $\langle 11-20 \rangle$  direction, as shown in **Fig. 4(c)**.

In order to investigate the feasibility of the selective growth of QWRs,  $\text{Al}_{0.24}\text{Ga}_{0.76}\text{N}/\text{GaN}$  layers were grown on the mesa structures using the optimum growth conditions obtained using planar substrates. **Figures 5(a) and 5(b)** show the cross-sectional SEM images of the samples after the growth of AlGaN layer on  $\langle 1-100 \rangle$ -oriented mesa structures with initial pattern sizes,  $W_0$ , of 750 nm and 200 nm, respectively. The grown thickness was 300 nm in terms of growth on a planar (0001) GaN substrate. After the growth, mesa structures having a top width,  $W$ , of 760 nm and 200 nm were obtained, whose values were equal to or larger than the initial pattern width,  $W_0$ . Furthermore, the angle of side facets measured with respect to a (0001) plane slightly decreased after growth, as shown in **Figs. 5(a) and 5(b)**.

On the other hand, it was clearly seen that the top width decreased after growth in the case of growth on the  $\langle 11-20 \rangle$ -oriented mesa. **Figures 6(a), 6(b) and 6(c)** show the cross-sectional SEM images of the samples after the growth of AlGaN layers on the  $\langle 11-20 \rangle$ -oriented mesa structures with initial pattern sizes,  $W_0$ , of 900 nm, 350 nm and 50 nm, respectively. The mesa top widths,  $W$ , of 820 nm and 240 nm were obtained, respectively, for the samples with  $W_0$  of 900 nm and 350 nm. As for the sample with  $W_0$  of 50 nm, a sharp

ridge structure with  $W$  below 10 nm was obtained after the growth. Furthermore, it was found that the angle of side facets remained constant at  $62^\circ$ , corresponding to the angle of (1-101) facets, during the growth for all samples, as seen in **Figs. 6(a), 6(b) and 6(c)**.

The difference between the initial top width of mesa,  $W_0$ , and the top width of mesa after the growth,  $W$ , is plotted in **Fig. 7** as a function of the grown thickness for two cases of growth on <1-100>- and <11-20>-oriented mesa stripes. In the case of the growth of the GaN layer,  $W$  decreased with growth thickness for both mesa orientations. The latter case indicates that the growth rate of GaN on the top facet is much larger than that on the side facet. This is very similar to the case of growth on GaAs (001) and (111)B patterned substrates [6].

Similarly, for growth of AlGaIn on <11-20>-oriented mesas, the top width decreased as a function of growth thickness. On the other hand, in the case of the growth of AlGaIn on <1-100>-oriented mesas,  $W$  increased with growth thickness. This is probably because the growth rate on the side facets of the <1-100>-oriented mesa became larger than that on the top (0001). Such behavior could not be observed in the growth of GaN on the <1-100>-oriented mesas. The present results indicate that, reflecting complex growth kinetics involving reaction and diffusion, incorporation rates of group-III adatoms are different depending on the adatom species, crystalline facets and growth conditions.

In order to further investigate how the wire width can be controlled, repeated wire growth experiments were carried out. **Figure 8** shows the cross-sectional SEM image of AlGaIn/GaN multilayers grown on the <11-20>-oriented mesa. The lateral width of GaN wires formed on the top facets was limited by two facet boundary planes (FBPs) which separate the growth region between the neighboring facets. The angle of FBPs,  $\theta$ , defined with respect to the (0001) plane is found to be  $\theta = 82^\circ$  according to **Fig. 8**. According to our previous paper [6],  $\theta$  should satisfy

$$\tan \theta = \frac{\alpha \cdot \tan \theta_{\text{side}} - \tan \theta_{\text{top}}}{\alpha - 1}, \quad (1)$$

where  $\theta_{\text{top}}$  and  $\theta_{\text{side}}$  are the angle of the top facet and that of the side facet with respect to the top facet of the mesa pattern, respectively, and  $\alpha$  is the ratio of the vertical growth rate on a top facet to that on a side facet. In the present case of the growth on <11-20>-oriented mesas, the values of  $\theta_{\text{top}} = 0^\circ$  and  $\theta_{\text{side}} = 62^\circ$  should be used as the angles of top (0001) facet and side (1-101) facets, respectively.

By using eq. (1) and the measured value of  $\theta = 82^\circ$ , the growth selectivity,  $\alpha$ , is estimated to be 1.35. According to our model for the evolution of facet boundary planes,  $W - W_0$  should be given by the following equation using the growth thickness on the top facet,  $t$ , and the boundary angle,  $\theta$ .

$$W - W_0 = -\frac{2t}{\tan \theta} \quad (2)$$

The lateral wire width geometrically estimated by this simple relation is also plotted in **Fig. 7** for the growth of AlGaIn on <11-20>-oriented mesas. It shows good agreements with the experimental data. Thus, the lateral wire width of a QWR structure formed selectively on the top of an AlGaIn mesa can be controlled by adjusting the growth thickness and the initial size of mesa patterns.

### 3.3 Selective growth of QWR structures

On the basis of the above growth selectivity measurements, the selective growth of AlGaIn/GaN QWR arrays was attempted on the <11-20>-oriented mesa substrates, using the material supply shown in **Fig. 3(b)**. **Figure 9** shows the CL spectra obtained from the wire

region of the arrays of QWR structures. After the growth of the AlGaIn/GaN/AlGaIn sandwiched layer, the top width of the mesa was reduced, as expected, maintaining a smooth surface morphology, as seen in the inset of **Fig. 9**. As shown in **Fig. 9**, two sharp emissions were observed at 3.49 eV and 3.69 eV, in addition to a broad yellow luminescence peak from GaN in the lower energy region.

The peak observed at 3.49 eV comes from the band emission of GaN. Another peak observed at a higher energy of 3.69 eV proves the successful formation of embedded AlGaIn/GaN QWR structures. However, evidence of lateral quantum confinements was not observed for the present sample. This is because the lateral wire width of the present wire structure, grown as a first attempt, is estimated from SEM observation to be 800 nm, which is too large to expect a lateral confinement. The value of 3.69 eV is consistent with the vertical confinement energy of 3.64 eV that is estimated from the supply thickness of the GaN QWR layer. By further reduction of the initial mesa width,  $W_0$ , and by further optimization of growth conditions, it seems that narrower QWRs with strong lateral confinements can be realized.

#### 4. Conclusions

In this study, the feasibility of the selective MBE growth of AlGaIn/GaN QWR structures on prepatterned substrates was investigated. The main conclusions are listed below.

- (1) In situ RHEED, XRD and SEM studies have revealed that the growth of GaN and AlGaIn in the present study takes place in the group-V limited mode, while maintaining crystal quality similar to that of the MOVPE GaN template.
- (2) The selective growth of AlGaIn/GaN QWR structures is possible on mesa patterns fabricated in a  $\langle 11\bar{2}0 \rangle$  orientation where the lateral wire width formed selectively on the top facet of the mesa can be controlled by adjusting the growth thickness and the initial size of mesa patterns.
- (3) From CL measurements, the emission from the embedded AlGaIn/GaN QWR structure has been clearly identified. Further optimization of growth conditions should lead to the realization of QWRs with strong lateral confinements that are useful for electronic and photonic device applications.

#### Acknowledgements

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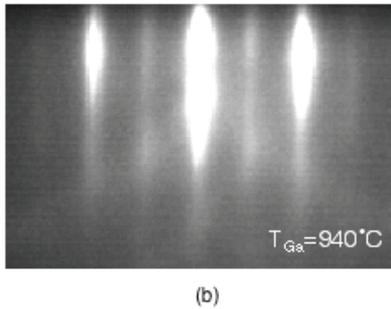
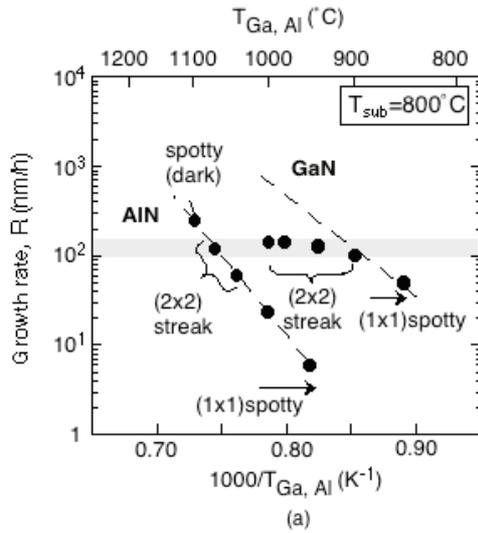


Fig. 1: (a) Growth rates of GaN and AlN and (b) (2x2) RHEED pattern obtained during GaN growth at  $T_{Ga} = 940^\circ\text{C}$ .

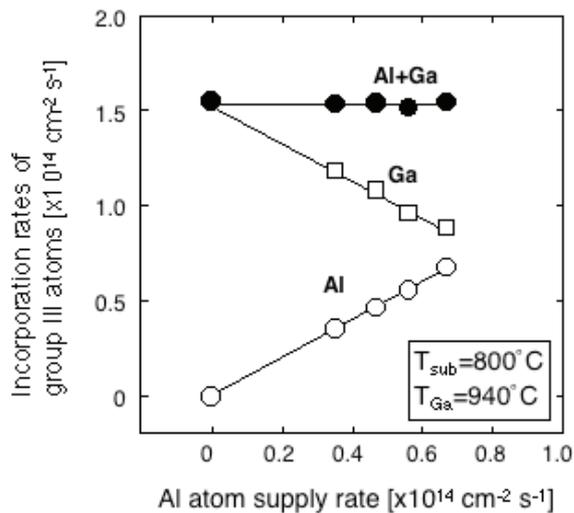


Fig.2: Incorporation rates of group-III atoms on planar substrates. The dashed lines indicate the growth rates expected from Arrhenius relationships of Ga and Al supplies from K-cells.

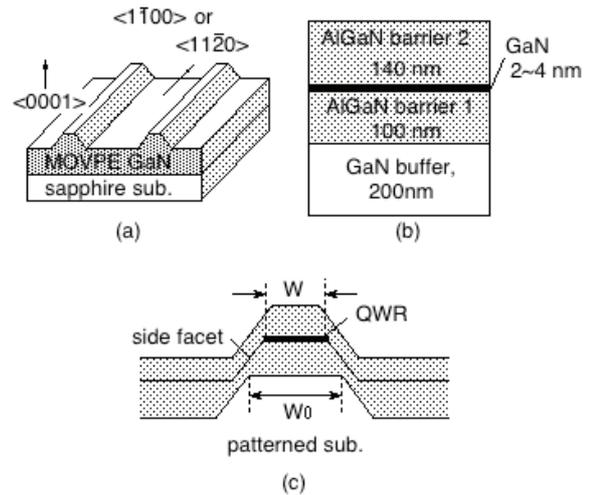


Fig. 3: (a) Patterned substrate, (b) material supply and (c) schematic illustration of embedded QWR.

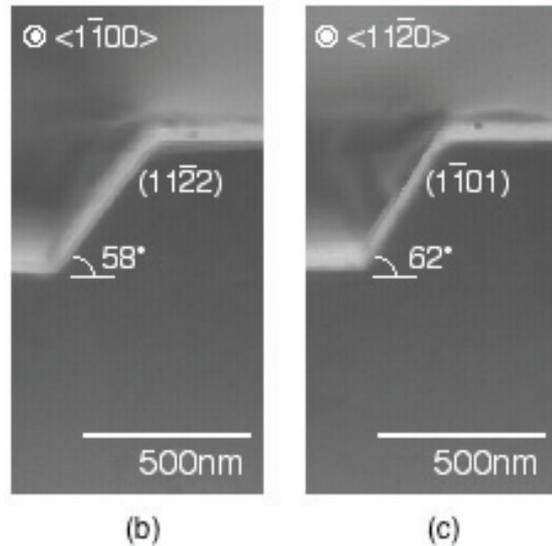
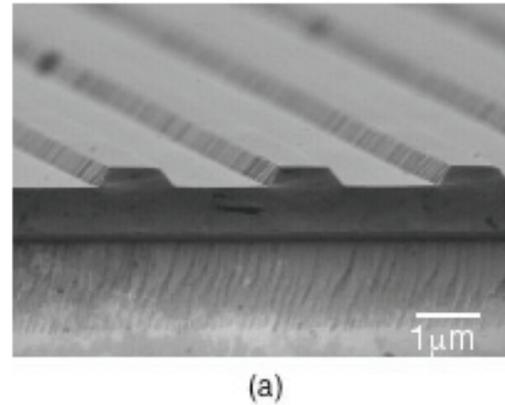
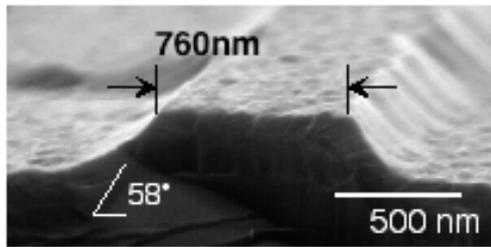
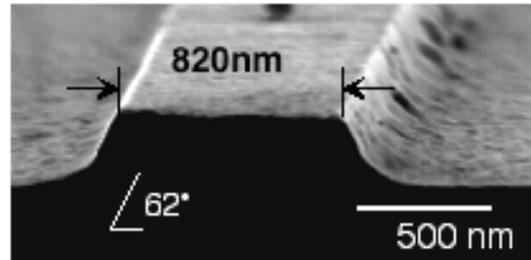


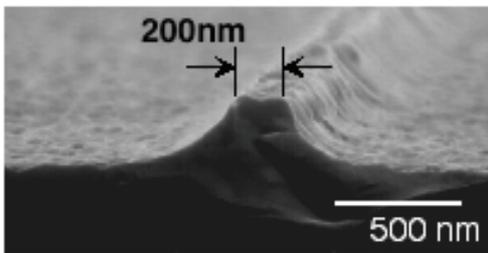
Fig. 4: Cross-sectional SEM images of patterned substrates fabricated by ECR-RIBE etching process.



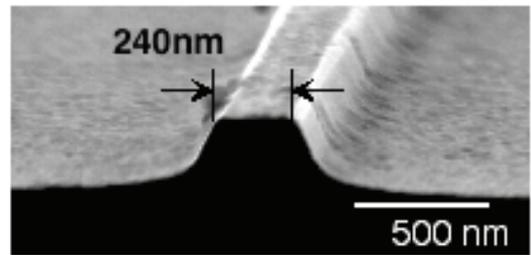
(a)



(a)

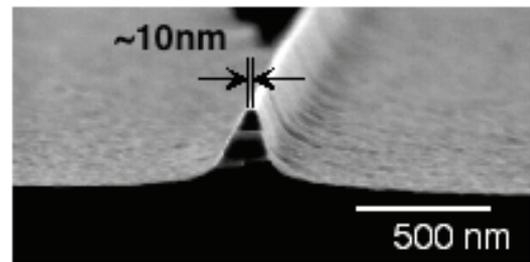


(b)



(b)

Fig. 5: Cross-sectional SEM images of samples after growth on  $\langle 1-100 \rangle$ -oriented mesas having different pattern sizes of (a)  $W_0=750$  nm and (b)  $W_0=200$  nm.



(c)

Fig. 6: Cross-sectional SEM images of samples after growth on  $\langle 11-20 \rangle$ -oriented mesas having different pattern sizes of (a)  $W_0 = 900$  nm, (b)  $W_0 = 350$  nm and (b)  $W_0 = 50$  nm.

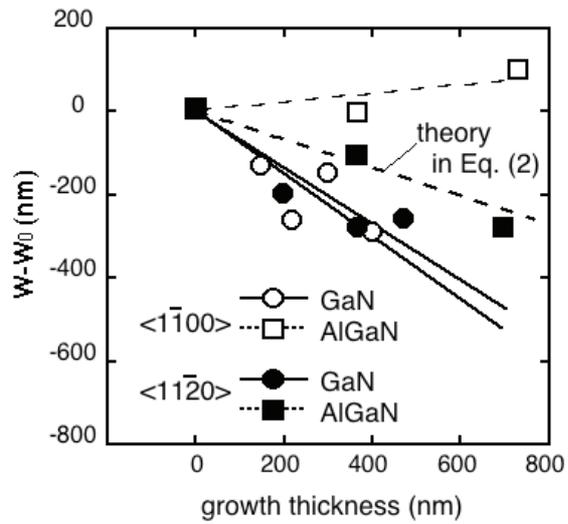


Fig. 7: Plot of top width of mesa vs. growth thickness.

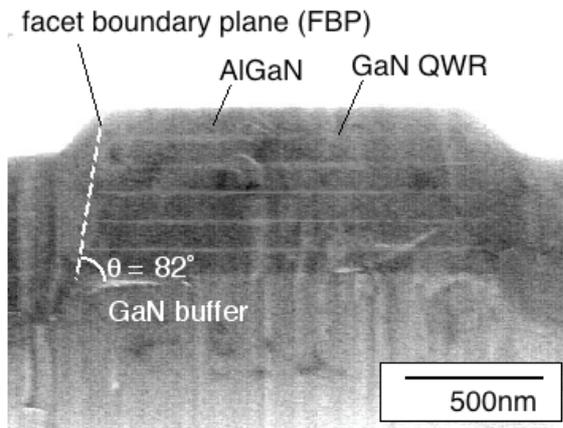


Fig. 8: Cross-sectional SEM images of GaN/AlGaN multilayer grown on  $\langle 11\bar{2}0 \rangle$ -oriented mesa.

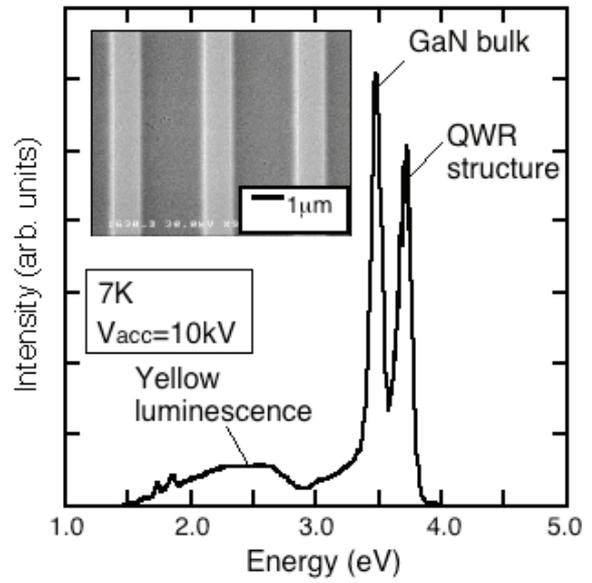


Fig. 9: CL spectra obtained from  $\langle 11\bar{2}0 \rangle$ -oriented QWR structure.