Study on ECR Dry Etching and Selective MBE Growth of AlGaN/GaN for Fabrication of Quantum Nanostructures on GaN (0001) Substrates

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Abstract
This paper attempts to form AlGaN/GaN quantum wire (QWR) network structures on patterned GaN (0001) substrates by selective MBE growth. Substrate patterns were prepared along <1120>- and <1100>-directions by ECR-assisted reactive-ion beam etching process. Selective growth was possible for both directions in the case of GaN growth, but only in the <1120>-direction in the case of AlGaN growth. A hexagonal QWR network was successfully grown on a hexagonal mesa pattern by combining the <1120>-direction and two other equivalent directions. AFM observation confirmed excellent surface morphology of the grown network. A clear CL peak coming from the embedded AlGaN/GaN QWR structure was clearly identified.

Keywords: GaN, AlGaN, quantum wire, selective MBE growth, dry etching
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1. Introduction
The AlGaN/GaN system seems to be useful, not only to blue/UV photonic and high-power electronic devices, but also to high temperature operating quantum devices because of wide energy gaps with a large $\Delta E_c$ and availability of high density 2DEG even under undoped conditions which avoid the doping fluctuation issues in nano-devices. Several groups [1-3] have reported the growth of GaN quantum dots using the anti-surfactant or Stranski-Krastanov (SK) effects. However, position- and size- controllability is rather poor in these growth modes. On the other hand, we have shown that the selective molecular beam epitaxy (MBE) growth on a pre-patterned substrate is a very powerful approach for formation of position- and size- controlled high density quantum nanostructures on GaAs and InP substrates [4, 5].

The purpose of this paper is to study and optimize ECR dry etching and MBE growth processes for GaN and AlGaN/GaN in order to realize a selective MBE growth method of nitride based quantum nanostructures. Key points in selective MBE growth are availability of a facet-revealing etching process for pattern preparation, and availability of suitable growth selectivity on different facets of the pattern for growth. These points were clarified through an attempt to grow hexagonal quantum wire (QWR) networks.

2. Experimental
GaN and Al$_x$Ga$_{1-x}$N (x=0-0.43) layers were grown on n-type (0001) GaN/sapphire templates by a RF-radical assisted MBE process. Templates were commercial MOVPE materials.
Both of uniform growth on planar substrates and selective growth on patterned substrates were investigated.

As templates for selective growth, straight mesa stripes and hexagonal nanowire network patterns, as shown in Fig. 1, were formed by etching of GaN/sapphire substrates. Since conventional wet chemical etching is difficult for nitride materials, etching was carried out by an electron cyclotron resonance assisted reactive ion beam etching (ECR-RIBE) process developed for nitrides by our group [6]. It uses a methane-based gas mixture of CH$_4$/H$_2$/Ar/N$_2$ = 5/15/3/3 sccm where addition of N$_2$ is extremely important. A typical etching rate was about 10 nm/min, and this was suitable for controlled fabrication of nanometer scale patterns. In addition, this process produced sidewalls corresponding to well-defined crystalline facets.

For MBE growth, nitrogen radicals were excited with a microwave power of 350W and the N$_2$ flow rate of 0.5sccm. Template surfaces were cleaned in organic solvents and treated in a HF solution for 6min. Then, the template surface was further cleaned thermally in the MBE chamber at 800°C for 5min in the N$_2$ radical atmosphere. Subsequently, an undoped GaN layer was grown at 800°C followed by growth of Al$_x$Ga$_{1-x}$N (x=0-0.43) single or multi-layers at the same temperature. The growth rate was 120 nm/h.

3. Results and Discussion

3.1 Growth of AlGaN on planar GaN template

According to RHEED observation, well-defined (2x2) streak patterns were observed and maintained during growth of both GaN and AlGaN layers.

Figure 2 shows the results of XRD measurements on the AlGaN layers grown at different supply rates of Al atom. Here, Ga source temperature, T$_{Ga}$, was fixed at 940°C. A minimum FWHM value of the XRD peak of 300 sec was obtained for both GaN and Al$_{0.24}$Ga$_{0.76}$N layers. This value was equal to or even smaller than that of the MOVPE GaN template used in this study.

The Al composition of Al$_x$Ga$_{1-x}$N layers was estimated by assuming the Vegard’s law, and the result indicated that it could be changed from x = 0.24 to 0.43 by changing Al source temperature, T$_{Al}$. However, for a fixed Ga source temperature and a fixed N supply, supply of Al beam did not change the total growth rate of the AlGaN layer. Namely, the Ga incorporation rate decreased with increase of Al supply, keeping the total group-III incorporation rate constant. This indicates occurrence of a group-V supply limited growth mode similarly to the case of InP growth using a polycrystalline InP source [7]. It also indicates that Al shows a higher incorporation rate than Ga during competitive incorporation.

3.2 Growth selectivity on stripe-patterned substrates

To investigate basic growth selectivity, <1120>- and <1100>-oriented mesa stripe patterns having with (0001) terraces were prepared on GaN (0001) substrates by the EB lithography and ECR dry etching. As a result, smooth stripes with a minimum terrace width (W$_t$) of 50nm were successfully realized for each direction. Each stripe had flat and smooth side facets. These features are suitable for the selective MBE growth of QWR structures [5]. An example of the cross-sectional SEM image of a fabricated substrate pattern is shown in Fig.3.

Growth on mesa stripes revealed the occurrence of growth selectivity whose behavior is complicated. Namely, selective growth was possible for both directions in the case of GaN growth, but only in the <1120>-direction in the case of AlGaN growth. To show the latter, the difference, W-W$_0$, of the top terrace width of the mesa before (W$_0$) and after (W) growth of an Al$_{0.24}$Ga$_{0.76}$N layer on GaN stripes, is plotted in Fig.4 vs. the grown thickness for two cases of growth on <11
00>-oriented and <11 2 0>-orientated stripes. The terrace width decreased with growth time on <11 2 0>-oriented GaN stripes whereas it increased on <1 1 00>-oriented GaN stripes. Obviously, the former is suitable for the QWR growth. Usui et al. [8] and Hiramatsu et al. [9] have reported that (1101) facets appear in <11 2 0>-oriented GaN stripes and that the growth rate on this facet is much slower than that on the (0001) terrace. The present result seems to be consistent with these reports.

3.3 Growth of hexagonal QWR network structure

Hexagonal QWR networks were grown by supplying AlGaN/GaN/AlGaN materials on hexagonal patterns combining <11 2 0>- , <1 2 1 0>- and <21 1 0>- orientations. The AFM image of the grown network is shown in Fig. 5 (a). The line-scan profiles along directions indicated in Fig. 5 (a) are given in Figs. 5 (b) and (c). Excellent surface morphology can be seen in all directions.

Figure 6 shows cathodoluminescence (CL) spectra obtained from the wire region. Two sharp emissions were obtained at 3.49eV and 3.74eV in addition to a broad yellow luminescence peak from GaN. The peak at 3.74eV proves a successful formation of embedded AlGaN/GaN QWR structures. The monochromatic CL image showed a spatially continuous hexagonal emission pattern, which showed smooth connections at vertices of hexagons.

4. Conclusion

In order to investigate the feasibility of selective MBE growth of nitride based quantum nanostructures, a basic study was made through an attempt to grow a hexagonal quantum wire (QWR) network. It is found that our ECR-RIBE dry etching process can produce patterns of acceptable quality, and that suitable growth selectivity can be obtained by MBE growth processes by selecting the pattern directions and growth conditions depending on the material. Preparing a hexagonal mesa pattern using <11 2 0>-directions, a uniform hexagonal QWR network was successfully grown.

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References

Fig. 1 Substrate patterns for (a) straight QWR array and (b) hexagonal network.

Fig. 2 XRD spectra obtained from Al$_x$Ga$_{1-x}$N layers grown by a rf-radical assisted MBE.

Fig. 3 A cross-sectional image of a GaN $<11\overline{2}0>$-oriented straight mesa-pattern.

Fig. 4 Plot of the top width of mesa vs. growth thickness.

Fig. 5 (a) an AFM image, (b) a line scan profile along line A and (c) line scan profiles along line B of the hexagonal network.

Fig. 6 CL spectra obtained from a hexagonal network (Vacc=5kV).