報告

ハロゲン系気相成長法によるGaN基板作製

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Self-separation of Freestanding GaN from Sapphire Substrates by Hydride Vapor Phase Epitaxy

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要旨

我々は新たに開発した自己分離法により高品質GaN基板の作製に成功した．GaN基板はハロゲン系気相成長方法により，サファイア基板上に成長し，サファイア／GaN界面にかかる熱応力と格子定数差により，結晶成長シーケンス中に自己分離させることにより作製した．作製したGaN基板のサイズは現在のところ23 mm x 22 mm，厚さは100-500 μmである．透過電子顕微鏡観察による結晶欠陥密度は10^6 cm^{-2} ~10^7 cm^{-2}と低く世界最高水準の品質であった．今後，従来のGaN系青色LEDに用いられていたサファイア基板をGaN基板に置き換えることにより更なる高性能化が期待できる．

Abstract

Freestanding GaN wafers were produced by a newly developed self-separation method. Thick GaN layers were grown using hydride vapor phase epitaxy on a sapphire substrate with GaN seeds. The separation of the thick GaN layers took place during the growth sequence at the interface of GaN/sapphire, because of thermal stress and lattice mismatch between GaN and sapphire. The size of the freestanding GaN wafers was 23 mm x 22 mm. The threading dislocation density at the top surface was 10^6 cm^{-2} ~10^7 cm^{-2}.

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1. Introduction

Nitride compound semiconductors are potential materials for high efficient optical devices in the ultraviolet to red spectral region. Excellent uniform epitaxial layers of GaN and nitride alloys were grown on sapphire substrates using low-temperature AlN buffer layer by metalorganic vapor phase epitaxy (MOVPE). 1)

One of the problems hindering progress in III-nitride devices is the lack of nitride substrates on which lattice-matched group III-nitrides can be grown. Usually, GaN epitaxial layers have been grown on sapphire substrates or SiC substrates. However, the difference of lattice constant and thermal expansion coefficient between these foreign substrates and GaN bring about deterioration of device performance, such as lifetime.

Homogeneous GaN epitaxial growth on GaN substrates will improve the performances of nitride light emitting devices compared to ones on sapphire substrates. Recently, there are several attempts to produce GaN substrates by separation from sapphire substrates.2)-5) Currently the largest freestanding GaN substrate has been obtained by the laser lift-off method.[5] In addition, there are other techniques such as using $NdGaO_3$ 6), 7) or GaAs 8) as substrates.

We have newly developed a self-separation method of freestanding GaN from sapphire substrates. Thick GaN layers have been grown on particular sapphire substrates by hydride vapor phase epitaxy (HVPE). The sapphire substrates have stripe shaped GaN seeds as shown in Fig. 1. The thick GaN layers separate from the sapphire substrates at the interface between GaN epitaxial layers and sapphire substrates during the growth sequence. The self-separation could be caused by thermal stress and lattice mismatch between GaN and sapphire. The narrow structure of the seeds makes the self-separation easy.

2. Experimental

The GaN seed layer and the AlN buffer layer were grown on (11-20) sapphire substrates with a horizontal metal-organic vapor phase epitaxy system under atmospheric pressure. The source gases are trimethylgallium, trimethylaluminium and ammonia (NH_3).

Each seed layer consisted of a GaN layer grown on an AlN buffer layer. Next, GaN seeds were formed by photolithography and reactive ion beam etching. The etching depth was extended into the sapphire substrate through the exposed GaN seed layer and the AlN buffer layer. After the masked material was removed, the particular GaN seeds remained on the sapphire substrates as shown in Fig. 1.

On the base substrate with the GaN seeds, a thick GaN layer was grown by conventional HVPE method under atmospheric pressure. Ga metal and NH_3 gas were used as the gallium and nitrogen sources, respectively. The Ga metal reacts with hydrogen chloride, forming GaCl. The GaCl was reacted with NH_3 to form GaN. Hydrogen gas was used as the carrier gas. The thick GaN layers separated spontaneously from the sapphire substrates during the growth sequence.

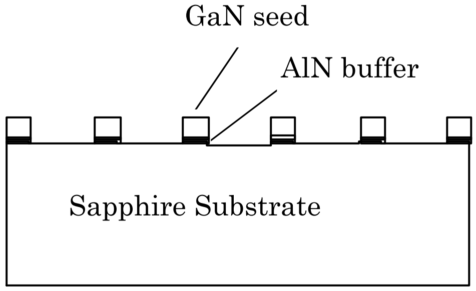

Fig. 1. Schematic diagram of substrate structure with GaN seeds

3. Result and Discussion

The morphology and defect microstructure were investigated using Normarski interference microscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM) and cathodoluminescence (CL).

Freestanding GaN wafers were successively grown as shown in Fig. 2. The size of the GaN wafer was 23mm x 22mm. The thickness of GaN wafers was 100 μm – 500 μm. Low magnification Normarski interference micrographs of the top surface are shown in Fig. 3. The surface is specular.

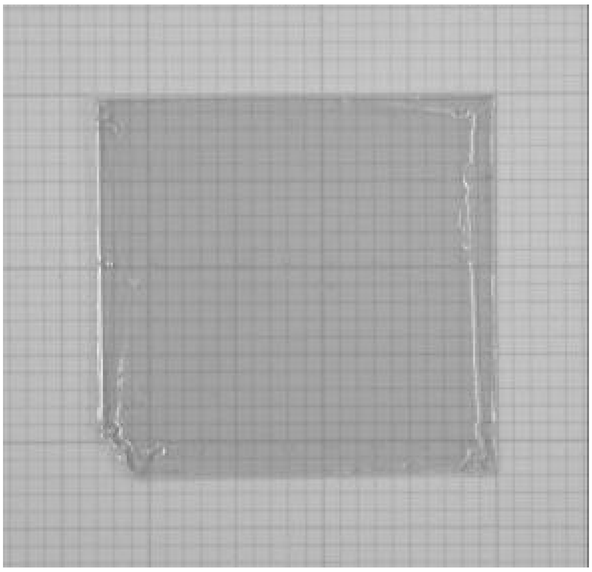

Fig. 2. Optical photograph of freestanding GaN wafers separated from sapphire substrates. Note grid elements are 1 mm x 1 mm

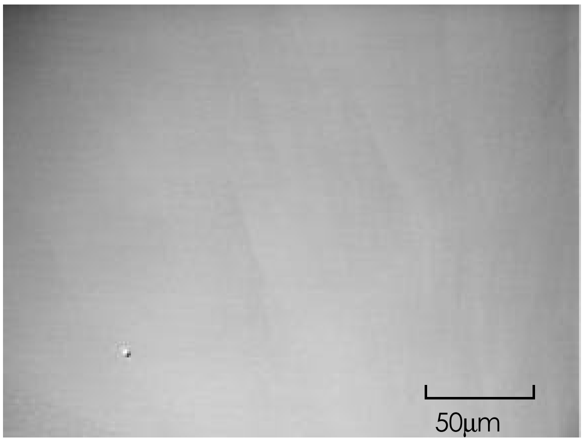

Fig. 3. Optical surface micrographs of GaN wafer separated from sapphire substrate

Figure 4(a) and 4(b) show cross-sectional TEM images of a GaN. Threading dislocations are not observed near the top surface in cross-sectional TEM image (Fig. 4(a)). The threading dislocation density was estimated to less than 10^7 cm^{-2} from the TEM image. On the other hand, figure 4(b) shows many threading dislocations that are not straight near the seed region of the GaN/sapphire interface. Many threading dislocations bend towards a lateral direction. The threading dislocation density was determined to about 10^9 cm^{-2} from the TEM image.

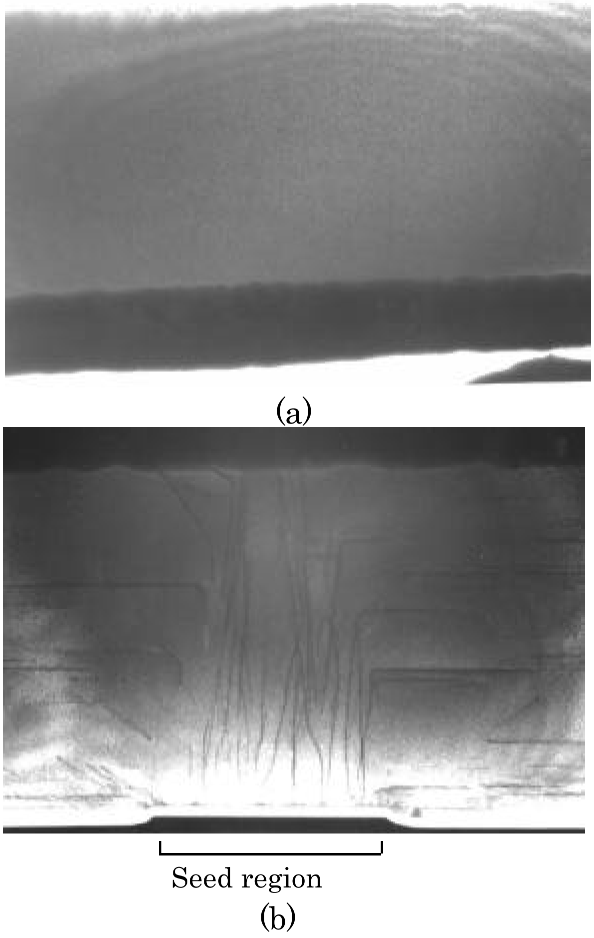

Fig. 4. Cross-sectional TEM images; (a) top surface region of a freestanding GaN wafer, and (b) bottom near the seed region of GaN/sapphire interface of a freestanding GaN wafer

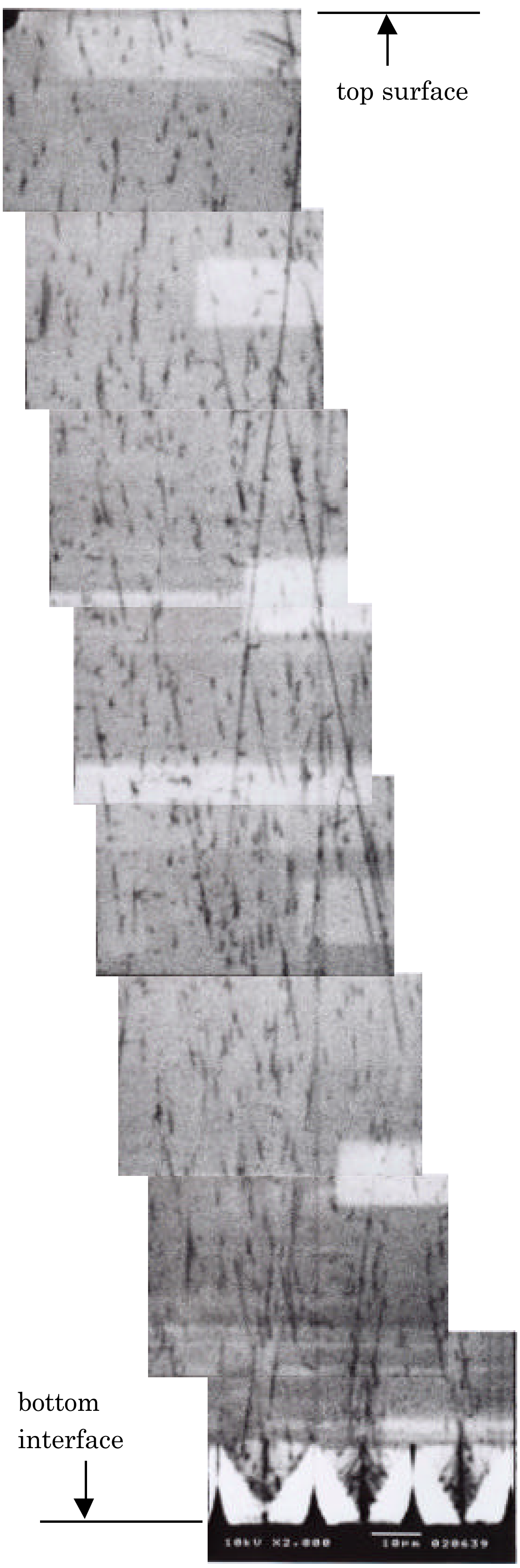

Fig. 5 Cross-sectional CL images from the top surface region to the bottom interface, including the seed region of GaN/sapphire interface of a freestanding GaN wafer

To investigate the behavior of the threading dislocations between the top surface and bottom surface in the GaN layers, we performed cross-sectional CL of the freestanding GaN. Figure 5 shows cross-sectional CL images from the top surface region to the bottom interface, including the seed region at the GaN/sapphire interface of a freestanding GaN wafer. The threading dislocations appear as dark spots and lines. In Fig. 6, variation of the dislocation density that was determined from this CL images is shown as a function of the distance from the top surface. The threading dislocation density does almost not change except in the facet growth region near the bottom. From these results, the reduction of threading dislocation density at the top surface region is believed to be due to dislocation bending at the early growth stage in GaN wafers.

We have also obtained GaN wafers of lower threading dislocation density, which is 1 x 10^6 cm^{-2}, from plan-view TEM observation.

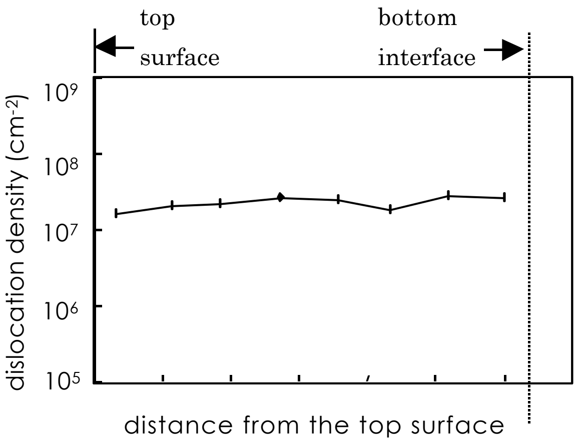

Fig. 6. Variation of dislocation density versus distance from the top surface by cross-sectional CL images

4. Conclusion

In conclusion, high quality freestanding GaN wafers have been successfully produced by the novel self-separation method. The thick GaN layer was grown on a sapphire substrate with a narrow structure of GaN seeds. The separation of the thick GaN layer took place during the growth process at the interface of GaN/sapphire, because of the lattice mismatch and the thermal stress between GaN and sapphire. The dislocation density of GaN wafers was about 10^6 cm^{-2} ~10^7 cm^{-2}.

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