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Heteroepitaxial Growth of GaN on γ-Al_2O_3/Si Substrate by Organometallic Vapor Phase Epitaxy

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Heteroepitaxy of GaN on both Si(001) and (111) substrates was investigated by atmospheric pressure organometallic vapor phase epitaxy. GaN layers on Si were deposited using an epitaxially grown intermediate layer of γ-Al_2O_3. When Si(001) was used as the substrate, highly oriented polycrystalline GaN was obtained. On the other hand, single-crystalline GaN layers could be obtained on Si (111) substrates with an epitaxial relationship of GaN(0001)/ γ-Al_2O_3(111)/Si(111) and GaN [2-1-10] // γ-Al_2O_3 [1-10] // Si [1-10]. GaN epilayers on Si(111) with a γ-Al_2O_3 intermediate layer indicated a (0002) X-ray rocking curve linewidth of 1000 arcsec and strong near band-edge photoluminescence without deep-level-related emission. The photoluminescence linewidth was comparable to that of GaN grown on sapphire substrate.

1. Introduction

Group-III nitrides have attracted much attention for use in optoelectronic devices operating in the ultraviolet to visible region, because the band gap can be varied from 6.2 eV for AlN to 1.9 eV for InN by changing the composition. Moreover, group-III nitrides have high potential for realizing high-power electron devices operating in the microwave region. Although bright light-emitting diodes (LEDs) and CW operation of laser diodes (LDs) have been successfully realized by using GaN layers grown on α-Al_2O_3 substrates, the poor thermal conductivity of α-Al_2O_3 leads to a serious problem regarding the thermal management of the devices, particularly during high power operation. In order to solve the

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heat sink problem, SiC, which has one of the highest thermal conductivties, has been used as the substrate for high-power device applications. However, it is difficult to grow high-quality bulk SiC because of its high synthesis temperature and the difficulty of the polytype controllability, and thus the SiC wafer becomes highly expensive.

Si is one of the most widely used semiconductors and has a relatively high thermal conductivity, high controllability of conduction type and carrier concentration, and high processibility, in addition to low cost. Moreover, it is possible to integrate GaN-related optoelectronic and/or high-power devices with Si integrated circuits, if high-quality GaN can be grown on Si. Therefore, Si has high potential for use as a substrate for GaN growth. In order to grow high-quality GaN layers on Si, various buffer layers, such as AlN,(1) SiC,(2,3) AlAs,(4) Ga_2O_3,(5) and Al_xO_y(6,7) have been investigated for preventing the formation of an amorphous SiN_x layer on the substrate surface, which occurs at temperatures even below 200°C.(8)

γ-Al_2O_3 has a relatively small lattice mismatch of $\Delta a/a \sim 2.4\%$ relevant to Si, and thus it has been proposed for the fabrication of a silicon-on-insulator (SOI) structure,(9) in which epitaxially grown γ-Al_2O_3 thin layers on Si are used as the insulator.(10) Since the γ-Al_2O_3 epilayer on Si substrate is stable in H_2 atmosphere even at a temperature of 1000°C, it can be utilized as an intermediate layer for GaN growth. Wang *et al.* used a γ-Al_2O_3 layer for GaN growth on Si(001) substrates, but the obtained GaN layer had a double domain structure, and a single crystalline GaN layer could not be obtained.(7)

We have investigated the growth of GaN on a γ-Al_2O_3(111)/Si(111) epi-wafer by molecular beam epitaxy (MBE) and demonstrated a single crystalline GaN layer.(11) However, details of the properties of grown films were not well investigated. In this study, we grow GaN on both Si(001) and Si(111) wafers by organometallic vapor phase epitaxy (OMVPE) using an epitaxially grown γ-Al_2O_3 intermediate layer, and investigate the properties of grown layers.

2. Experimental Details

GaN was grown on both Si(001) and Si(111) wafers using an epitaxially grown γ-Al_2O_3 intermediate layer by means of atmospheric pressure OMVPE. γ-Al_2O_3 epi-layers were grown by ultrahigh-vacuum chemical vapor deposition (UHV-CVD) and/or molecular beam epitaxy (MBE). Details of the growth conditions for γ-Al_2O_3 have been described in previous papers.(12–14) The nominal layer thickness of γ-Al_2O_3 was set to be in the range of 3–10 nm. After the growth of γ-Al_2O_3, the wafers were transferred to the OMVPE system. Using the conventional two-step growth method, in which a low-temperature-grown GaN buffer layer was used as the nucleation layer and to accommodate the lattice mismatch, we carried out the epitaxial growth of GaN. The growth conditions are summarized in Table 1, and the growth sequence is illustrated in Fig.1.

The crystalline quality of the grown GaN layer was examined by reflection high-energy electron diffraction (RHEED), X-ray diffraction (XRD), a Nomarski interference microscope, an atomic force microscope (AFM), and photoluminescence (PL) measurements.

Table 1
Growth conditions for GaN epitaxy on γ-Al_2O_3/Si.

	Thermal cleaning	GaN buffer	GaN epilayer
Substrate temperature [°C]	1000–1050	550	930–1000
Time [min]	5	2	120
TMGa flow rate [mmol/min]	–	18	18
V/III ratio	–	5000	5000
Carrier gas	H_2	H_2+N_2	H_2+N_2

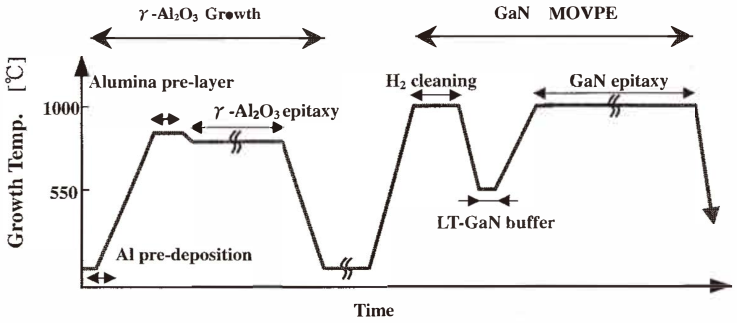
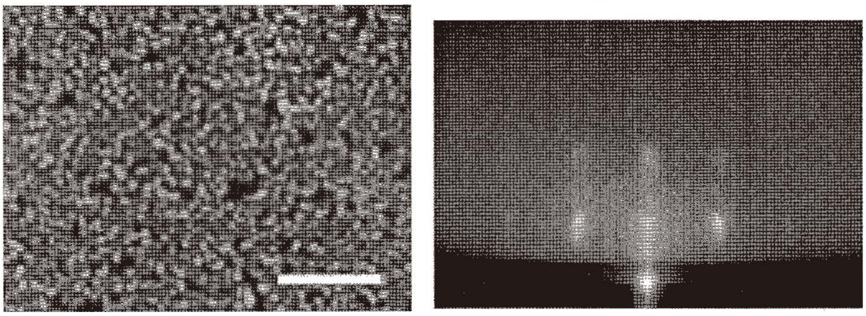


Fig. 1. Schematic diagram of the growth sequence for the GaN layer on Si substrate using epitaxially grown thin γ-Al_2O_3 layer.

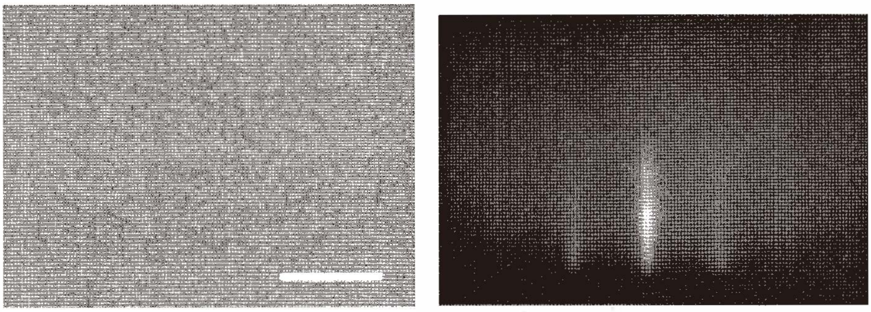
3. Results and Discussion

First, the crystalline quality of the γ-Al_2O_3 epilayer on the Si substrate was investigated. Figure 2 shows RHEED and AFM images of the as-grown γ-Al_2O_3 layer on (a) Si(001) and (b) Si(111) substrates. In the case of the Si(001) substrate, the RHEED pattern shows a clear spot pattern with weak streaks, which means that the γ-Al_2O_3 layer is single-crystalline. From the AFM observation it is seen that the γ-Al_2O_3 layer has a columnar structure of sub-nanometer scale, and the root-mean-square (rms) surface roughness evaluated in the area of 2 μm×2 μm is about 3.6 nm. On the other hand, the γ-Al_2O_3 layer grown on Si(111) presents a streak RHEED pattern and a very smooth surface with the rms surface roughness of 0.3 nm. The results indicate that both the crystalline quality and the surface flatness are better for γ-Al_2O_3 /Si(111). The stability of the γ-Al_2O_3 epilayer on the Si substrate in H_2 and NH_3 around 1000°C was investigated. When the H_2 and/or NH_3 treatment temperature is below 1000°C, no significant degradation can be seen on the γ-Al_2O_3 epilayer treated in both H_2 and NH_3, but pyramidal shaped pits occur above 1050°C. In order to obtain an atomically flat γ-Al_2O_3 layer, we used an alumina pre-layer, which is formed by a solid reaction between the SiO_x protective layer on the Si and metallic Al. These pits would be caused by thermal etching of the Si-rich region in the alumina pre-layer appearing on top of the γ-Al_2O_3 epilayer, because the layer thickness of γ-Al_2O_3 used



AFM Image RHEED E.B.//Si[100]

(a) γ–Al$_2$O$_3$/Si(001)



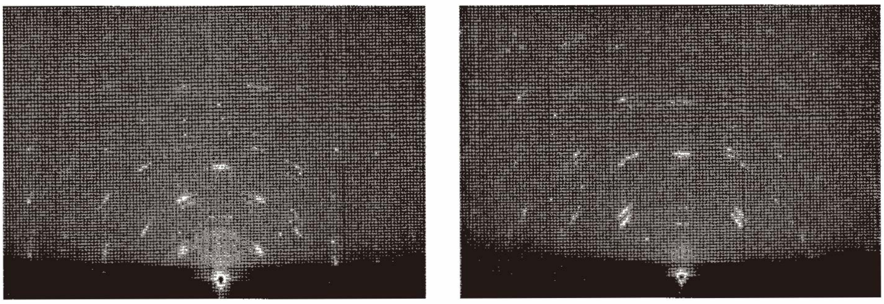
AFM Image RHEED E.B.//Si[110]

(b) γ–Al$_2$O$_3$/Si(111)

Fig. 2. AFM and RHEED images of as-grown γ- Al$_2$O$_3$ layer on a Si(001) and (b) Si(111) substrates. Marker in the AFM image represents 0.5 μm and the Z-range is 10 nm.

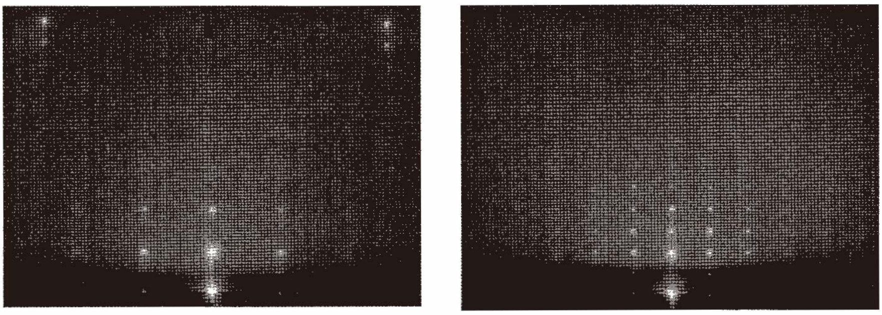
in the present work was very thin (nominally <10 nm). According to the results, the thermal cleaning temperature in H_2 was set to be 1000°C.

Figure 3 shows the RHEED pattern of GaN layers grown on (a) γ-Al$_2$O$_3$(001)/Si(001) and (b) γ-Al$_2$O$_3$(111)/Si(111). In the case of GaN grown on a γ-Al$_2$O$_3$(001)/Si(001) substrate, the RHEED pattern indicates that the GaN is a highly oriented polycrystalline layer. The major crystallographic orientation seems to be GaN(10-10)/γ-Al$_2$O$_3$(001) and GaN(0001)/γ-Al$_2$O$_3$(001), where the epitaxial relation of GaN(0001)/γ-Al$_2$O$_3$(001) is the same as that reported by Wang *et al.*(7) In order to determine the reason for the obtained layer structure, the growth processes were traced. In the growth of the GaN buffer layer, the buffer layer is polycrystalline, and remains polycrystalline after being heated to the epitaxial growth temperature. This growth process is quite different from that of GaN/α-Al$_2$O$_3$(0001), in which the GaN buffer is single crystalline just before the epitaxial growth.



E.B.//Si[110] E.B.//Si[100]

(a)GaN/γ–Al$_2$O$_3$/Si(001)



E.B.//GaN[10–10] E.B.//GaN[11–20]

(b)GaN/γ–Al$_2$O$_3$/Si(111)

Fig. 3. RHEED pattern of GaN epilayer grown on (a) Si(001) and (b) Si(111) with γ-Al$_2$O$_3$ layer at the epitaxial temperature of 975°C.

The reason for this difference is the rough surface of the γ-Al$_2$O$_3$ epilayer on Si(100), i.e., a part of the GaN nuclei are formed at the sidewall of the γ-Al$_2$O$_3$ column because of the relatively rough surface. Since in GaN there is a strong tendency for c-axis oriented growth, the c-axis of GaN nuclei formed on the sidewall of the column would be different from that for nucleation on top of the column. Therefore, it is expected that we can obtain single-crystalline GaN on Si(001) using γ-Al$_2$O$_3$ epitaxial intermediate layer by improving the surface morphology of γ-Al$_2$O$_3$ (001).

On the other hand, when the γ-Al$_2$O$_3$(111)/Si(111) wafers are employed, single crystalline GaN can be obtained. The epitaxial orientation relationship determined by RHEED and XRD is GaN(0001)/γ-Al$_2$O$_3$(111)/Si(111) and GaN [2-1-10] // γ-Al$_2$O$_3$ [1-10] // Si [1-10]. The lattice mismatch between GaN and g-Al$_2$O$_3$ is in the range of 10–13%, which depends on the direction, because γ-Al_2O_3 is a defective spinel structure and the surface atom position of (111) is not symmetrical. The surface morphology of the GaN on Si(111) is still rough under the Nomarski interference microscope observation, as can be expected from the spotty pattern of RHEED in Fig. 3.

Figure 4 shows (a) $2\theta/\omega$-mode and (b) ω-mode XRD scans for the GaN layer grown on

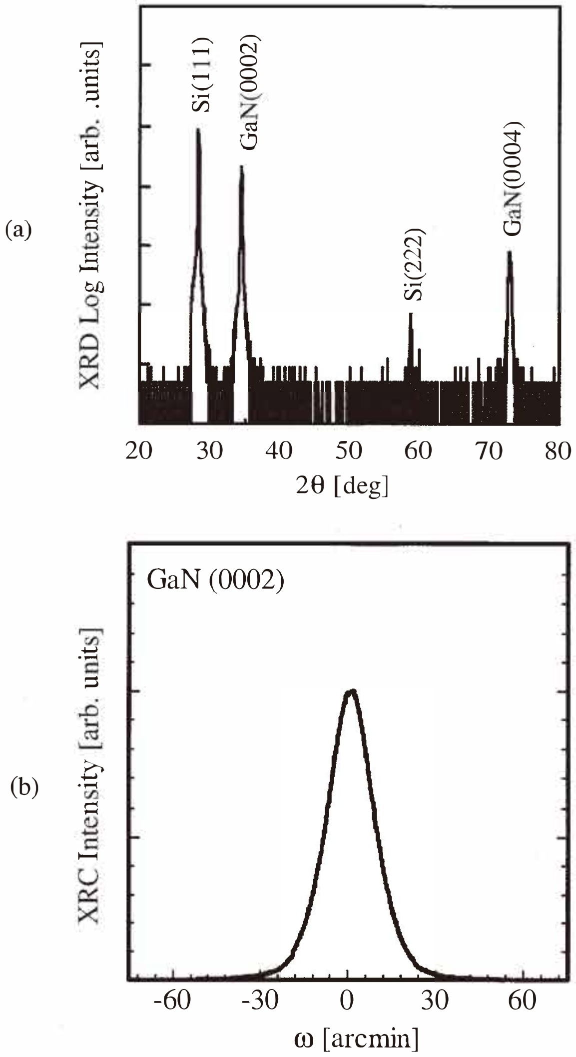


Fig. 4. X-ray diffraction profile of (a) $2\theta/\omega$ scan and (b) ω-scan for the 1.6-μm-thick GaN on γ-Al_2O_3/Si(111).

γ-Al_2O_3/Si(111). The full-width at half maximum (FWHM) of ω-mode ($\Delta\omega$) scans of (0002) reflection for a 1.6-μm-thick GaN layer was 1000 arcsec, which is wider than that for the GaN layer grown on α-Al_2O_3 (0001) substrates but much narrower than that for the GaN on γ-Al_2O_3(001)/Si(001).(7)

Figure 5 shows a typical PL spectrum of a GaN layer on a γ-Al_2O_3/Si(111) substrate compared with that of GaN grown on α-Al_2O_3(0001). In the figure, strong emission can be clearly seen in GaN/γ-Al_2O_3/Si(111) as well as GaN/α-Al_2O_3(0001), and the intensity of the yellow luminescence is very weak. It is difficult to compare the PL intensity between GaN/γ-Al_2O_3/Si and GaN/α-Al_2O_3 structures directly, because a part of the emitted light is incident into the Si substrate and absorbed, however the line width of the near-band-edge emission of GaN on the γ-Al_2O_3/Si substrate is comparable to that of GaN grown on α-Al_2O_3. Therefore, the optical quality of the GaN layer grown on γ-Al_2O_3/Si(111) wafers is comparable to that of a GaN layer grown on α-Al_2O_3. The peak position of the GaN on γ-Al_2O_3/Si wafers shifts to longer wavelength at about 16 meV. The linear thermal-expansion coefficients for α-Al_2O_3, GaN, and Si are 7.5×10^{-6}, 5.6×10^{-6}, and 4.1×10^{-6} K^{-1}, respectively. Therefore, the GaN on α-Al_2O_3 has a compressive strain caused by the difference of the thermal expansion between the sapphire and GaN, while the GaN layer on Si has a tensile strain. The difference of the residual stress on the GaN layer leads to the redshift of the PL peak position. However, the preliminary result of the FWHM of the ω-mode scan is comparable to that of growth with an AlN buffer layer,(15) and the γ-Al_2O_3 epitaxial intermediate layer has a high potential for the heteroepitaxy of GaN on Si(111) substrates. Further improvements can be achieved by investigating the initial growth stage of GaN/γ-Al_2O_3 and optimizing the growth conditions.

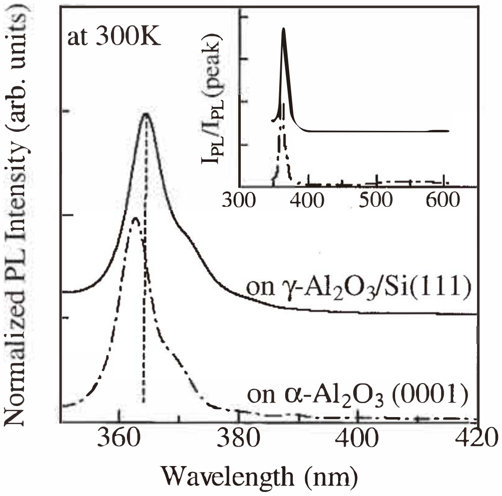


Fig. 5. Photoluminescence spectra of GaN grown on γ-Al_2O_3/Si compared with that of GaN grown on α-Al_2O_3 substrate.

4. Conclusions

GaN layers have been grown on both Si(001) and (111) substrates with a γ-Al_2O_3 thin epilayer by organometallic vapor phase epitaxy. Reflection high-energy diffraction and atomic force microscope measurements revealed that γ-Al_2O_3 was thermally stable in both H_2 and NH_3 up to 1000°C, and was effective for preventing the formation of SiN_x on the Si surface. Single crystalline GaN layers were obtained on γ-Al_2O_3(111)/Si(111). The orientation relationship was GaN(0001)/γ-Al_2O_3(111)/Si(111) and GaN[11-20] // γ-Al_2O_3[-110] // Si[-110], in which the lattice mismatch between GaN and γ-Al_2O_3 was approximately 10–13%. Although no optimization of the growth conditions was made, the line width of the (0002) X-ray rocking curve of 1000 arcsec was obtained. The optical quality of the GaN epilayer was comparable to that of GaN grown on α-Al_2O_3. The use of the γ-Al_2O_3/Si(111) epitaxial wafer offers the possibility of a large-area substrate for GaN growth.

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