($\bar{2}01$) β -Gallium Oxide substrate for high quality GaN materials

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Abstract

($\bar{2}01$) oriented β -Ga₂O₃ has the potential to be used as a transparent and conductive substrate for GaN-growth. The key advantages of Ga₂O₃ are its small lattice mismatches (4.7%), appropriate structural, thermal and electrical properties and a competitive price compared to other substrates. Optical characterization show that GaN layers grown on ($\bar{2}01$) oriented β -Ga₂O₃ are dominated by intense bandedge emission with a high luminescence efficiency. Atomic force microscopy studies show a modest threading dislocation density of $\sim 10^8$ cm⁻², while complementary Raman spectroscopy indicates that the GaN epilayer is of high quality with slight compressive strain. Room temperature time-findings suggest that the limitation of the photoluminescence lifetime (~ 500 ps) is due to nonradiative recombination arising from threading dislocation. Therefore, by optimizing the growth conditions, high quality material with significant optical efficiency can be obtained.

Keywords: GaN, Ga₂O₃, LED, high efficiency, optical properties

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Recently, wide band gap (4.8 eV) Gallium Oxide (Ga_2O_3) attracted significant attention due to its unique electrical and optical properties, which make it suitable for many applications, such as sensors,¹ lasers,² solar cell³ and field-effect⁴ devices. Ga_2O_3 is characterized by its high conductivity, transparency and low lattice mismatch with GaN materials, which make it an ideal substrate for GaN. Recently, we found that (-201) $\beta\text{-Ga}_2\text{O}_3$ substrate has a significant potential to produce high efficiency GaN materials for laser diode (LD) and light emitting diode (LED) devices.⁵ The epitaxial orientation relationships between $\beta\text{-Ga}_2\text{O}_3$ and the GaN thin film are defined by $(010) \beta\text{-Ga}_2\text{O}_3 \parallel (11\text{-}20) \text{GaN}$ and $(\bar{2} \ 0 \ 1) \beta\text{-Ga}_2\text{O}_3 \parallel (0001) \text{GaN}$, resulting in a lattice mismatch of $\sim 4.7\%$.⁵ Other substrates that have been used for production of these materials, such as sapphire (Al_2O_3), SiC or Si, introduce several limitations. For example, Al_2O_3 has a high lattice mismatch (14%), which leads to high dislocation densities (TDDs),⁶ and low conductivity, whereas SiC is expensive and has lack of transparency.⁷ However, high efficiency optical GaN devices require a low lattice mismatch substrate, characterized by good transparency and conductivity properties.

In this paper, we show that the optical properties of a high-quality GaN epilayer grown on monoclinic $\beta\text{-Ga}_2\text{O}_3$ by metal organic chemical vapor deposition (MOCVD) that can meet the aforementioned criteria.

A $0.48\mu\text{m}$ thick AlN buffer layer was grown on $(\bar{2}01)$ -oriented $\beta\text{-Ga}_2\text{O}_3$ using AlN buffer layer by metal organic chemical vapor deposition (MOCVD) from Tamura Corporation. The MOCVD growth conditions are detailed in Ref.5. A commercial (Lumilog) GaN film with a low TDD ($4 \times 10^7 \text{ cm}^{-2}$) grown on sapphire ($\text{GaN}/\text{Al}_2\text{O}_3$) by MOCVD was used to compare the sample quality.

The surface morphology, roughness and microstructural defects of the epilayers were investigated by Atomic Force Microscopy (AFM) using a scanning probe microscope. Figure 1 shows the stepped surface morphology of the GaN/ Ga_2O_3 sample with atomic terraces $\sim 0.2 \text{ nm}$ in height. The root mean square (RMS) roughness is $\sim 0.68 \text{ nm}$, which indicates a very smooth surface compared to the commercial GaN/sapphire reference sample (with an RMS roughness of $\sim 0.87 \text{ nm}$). Termination of TDs in the epilayer surface is indicated by small pits in the AFM scan (Figure 1). The average density of TDs is found to be $\sim 10^8 \text{ cm}^{-2}$. In the pertinent literature, the TDD of GaN epilayers grown on sapphire typically varies in the $10^8\text{-}10^{10} \text{ cm}^{-2}$ range^{8,9} and our reference sample shows a low TDD of $4 \times 10^7 \text{ cm}^{-2}$. We also observed

presence of mixed dislocations (medium pits) and edge dislocations (smallest pits) at a ratio of 2:3 per μm^2 . These results are in agreement with the low x-ray diffraction rocking curve width of the GaN/Ga₂O₃ and GaN/Al₂O₃ epilayers, which confirms that, with typical values for state-of-the-art films, both samples are of good quality⁵

Room temperature (RT) Raman measurements were carried out in the Horiba LabRam ARAMIS micro-Raman system with backscattering geometry. The samples were excited by a 473 nm diode laser and the data were collected by a monochromator equipped with 1800 lines/mm grating and a CCD camera cooled by liquid nitrogen (Agilent 5400). Figure 2 shows the Raman spectrum of the GaN/ β -Ga₂O₃ epilayer and that of the β -Ga₂O₃ substrate, which serves as a reference for identifying the GaN peaks. The Raman peaks of the β -Ga₂O₃ substrate are marked by asterisks.¹⁰ However, we observe that all the translational modes of bulk β -Ga₂O₃ are redshifted in the GaN/Ga₂O₃ epilayer by 1.5 to 2 cm^{-1} when compared to bulk β -Ga₂O₃. It is expected that Raman active modes (one A_1 , one E_1 and two E_2) of GaN would be observed in the GaN/ Ga₂O₃ epilayer.¹¹ However, Figure 2 shows E_2 (high) (at 569.04 cm^{-1}) and E_2 (low) modes (at 143.2 cm^{-1}) only. The E_2 (high) mode peak is clearly observed. The symmetry E_2 (low) mode partially overlaps with the 144.4 cm^{-1} substrate mode (Ga₂O₃). The detected E_2 modes verify the backscattering through the c-axis of wurtzite GaN.¹² We observe that, relative to the positions of the peaks for GaN grown on sapphire, the positions of these peaks are dependent on the substrate due to different strain states for different samples.¹¹ The E_2 (high) mode has been demonstrated to be sensitive to biaxial strain in GaN epilayers.¹¹ Here, we posit presence of a slight compressive strain because the peak exhibits a blueshift of 1.04 cm^{-1} compared with that of bulk GaN (568 cm^{-1}).¹² These results agrees with the XRD analysis.⁵ In contrast, the Raman peaks of β -Ga₂O₃ experience a redshift, which indicates a corresponding tensile stress induced in the interface. In addition, the A_1 symmetry LO-phonon mode near 735 cm^{-1} is not observed in the Raman spectrum of GaN/Ga₂O₃, which may be due to the strong coupling between the A_1 mode of the LO-phonon and the carrier plasmon (LO-phonon-plasmon coupling modes of electronic excitation and lattice vibrations).¹³ Therefore, the LO-phonon-plasmon coupling causes the Raman peaks to be broader than those of a pure LO mode.¹⁴ Furthermore, no Raman peaks related to the cubic GaN structure were observed. Therefore, the Raman measurements confirm the XRD finding that the GaN epilayer is a single-phase wurtzite, and thus a high-quality material.⁵

Photoluminescence (PL) was measured to investigate the optical properties of the GaN film using a 325 nm cw He-Cd laser. The spectra were detected by an Andor monochromator attached to a CCD camera. The samples were mounted in a closed-cycle Helium cryostat for low-temperature PL (6 K). Figure 3(a) and 3(b) show RT and low temperature PL spectra of GaN/Ga₂O₃ and commercial GaN/Al₂O₃, respectively. The RT PL spectrum of GaN/ β -Ga₂O₃ is characterized by a dominant intensity near the bandedge emission centered at 3.41 eV and a very weak yellow luminescence emission. The bandedge intensity of GaN/Ga₂O₃ wafer is 1.5 times stronger at RT than that from the commercial GaN/Al₂O₃ wafer, although the TDD of the commercial GaN/Al₂O₃ is lower. This suggests that other defects that create a high density of nonradiative deep states (such as point defects or other structural defects) are significantly suppressed, resulting in much higher luminescence efficiency. Surprisingly, the ratio of PL intensity of the bandedge emission at 6 K to that at 300 K (2.7:1), is significantly lower than that of the commercial GaN/Al₂O₃ reference sample (20:1), which is similar to the value reported in literature.¹⁵ However, the full width half maximum (FWHM) of the GaN/Ga₂O₃ bandedge peak is larger than that measured for the sample grown on Al₂O₃. The redshift of the bandedge peak of GaN/Ga₂O₃ can be due to the slighter compressive strain of the sample compared to that of GaN/Al₂O₃.

RT PL time-resolved (RT-TR) spectroscopy was investigated by using Ti-Sapphire lasers attached to a third-harmonic generator, allowing 266 nm excitation wavelength to be used. The spectra were acquired by a Hamamatsu universal streak camera, with 76 MHz repetition rate and 22.5 mW excitation power. Figure 4 shows the RT-TR signal for both samples. The RT-TR signal pertaining to the GaN/Al₂O₃ sample exhibits one PL decay ($\tau = 719$ ps), whereas biexponential decay is shown for GaN/Ga₂O₃ $\tau_1 = 240$ ps, and $\tau_2 = 500$ ps. The biexponential behavior in GaN epilayer is a sign of recombination events occur through multilevel process.¹⁶ These findings may explain the broadening of the bandedge peak pertaining to this sample compared to that grown on sapphire. In addition, the PL lifetime of the GaN/Ga₂O₃ is shorter than that of GaN/Al₂O₃. Usually, the slow decay of GaN material is affected by the nonradiative recombination near TD.^{15,17} As nonradiative recombination occurs due to the carriers trapped near the TD, which results in short lifetime, the shorter decay time of GaN/Ga₂O₃ is, therefore, due to the higher TDD of this sample compared to the one grown on sapphire. Therefore, we find this result promising, as reducing the dislocation density by optimizing the growth of these samples can result in high optical efficiency GaN materials with significantly longer lifetime.

In conclusion, a high-quality GaN epitaxial thin film with high optical efficiency was grown on β -Ga₂O₃ substrate by MOCVD even under unoptimized growth conditions. The PL results indicated that the IQE was high (at about 37%) and the peak intensity at room temperature was considerably higher than that of the commercial GaN/Al₂O₃ reference sample. Time-resolved spectroscopy indicates that the effects of the high TD and the lifetime can be mitigated significantly by reducing the TDD. Therefore, β -Ga₂O₃ is a promising substrate for the growth of high-quality GaN films under systematically optimized growth conditions. This approach can yield highly efficient UV LEDs and LDs, due to high transparency and conductivity of β -Ga₂O₃ substrates.

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Figure 1. $1000 \times 1000 \text{ nm}^2$ AFM image of the GaN/Ga₂O₃ epilayer.

Figure 2. The RT Raman spectra measured from the GaN/Ga₂O₃. (Asterisks mark shows the signals from the β -Ga₂O₃).

Figure 3 The PL spectra at (a) 6 K and (b) 300 K, showing near bandedge emission and no noticeable yellow band.

Figure 4. the normalized luminescence of the RT-RT PL signal at the bandedge emission peak of each sample.