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Characteristics of atomic layer deposited Gd_2O_3 on n-GaN with an AlN layer

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The interfacial and electrical properties of atomic layer deposited Gd_2O_3 with an AlN layer on n-GaN were investigated. According to X-ray photoelectron spectroscopy spectra, the formation of Ga–O bonds that is significant near the $\text{Gd}_2\text{O}_3/\text{GaN}$ interface was suppressed near the AlN/ $\text{Gd}_2\text{O}_3/\text{GaN}$ and $\text{Gd}_2\text{O}_3/\text{AlN}/\text{GaN}$ interfaces. Larger amounts of oxygen atoms across the dielectric layers were observed for AlN/ $\text{Gd}_2\text{O}_3/\text{GaN}$ and $\text{Gd}_2\text{O}_3/\text{AlN}/\text{GaN}$ junctions, which in turn produced the dominant peak corresponding to O–Al bonds. The flatband voltage shift in capacitance–voltage hysteresis characteristics was highest for the $\text{Gd}_2\text{O}_3/\text{AlN}/\text{GaN}$ junction, indicating the highest interface and oxide trap densities. In addition, AlN/ $\text{Gd}_2\text{O}_3/\text{GaN}$ and $\text{Gd}_2\text{O}_3/\text{AlN}/\text{GaN}$ junctions showed the highest interface state densities in the energy ranges of 0.1–0.2 eV and 0.4–0.6 eV, respectively. The reverse leakage currents were explained by Fowler–Nordheim (FN) for $\text{Gd}_2\text{O}_3/\text{GaN}$ and AlN/ $\text{Gd}_2\text{O}_3/\text{GaN}$ junctions and by trap assisted tunneling (TAT) for the $\text{Gd}_2\text{O}_3/\text{AlN}/\text{GaN}$ junction.

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1. Introduction

Among rare earth (RE) oxide based materials, gadolinium oxide (Gd_2O_3) has been considered as a promising material for application in next-generation complementary metal–oxide–semiconductor (CMOS) transistors, because Gd_2O_3 has a wide band gap (~ 6.0 eV) with permittivity values of 10–14 for the epitaxial cubic phase, optimum band offset values with respect to Si, and high thermodynamic stability.^{1–3} Gd_2O_3 has also been considered as a gate oxide in Ge-based MOS devices⁴ and as a passivation layer for GaAs-based electronic devices.⁵ Upon subsection to a forming gas treatment, the atomic layer deposition (ALD) grown Gd_2O_3 layer showed promising dielectric behavior, with no hysteresis and reduced interface trap density.⁶

Plasma-enhanced chemical vapor deposition (PECVD)-grown SiN_x has been used as an effective passivation material to suppress the current collapse in AlGaIn/GaN metal–insulator–semiconductor heterostructure field-effect transistor (MIS-HFET).⁷ High- k dielectric oxides such as Al_2O_3 and HfO_2 have also been employed as a passivation layer in AlGaIn/GaN high electron mobility transistors (HEMTs).^{8,9} However, it was shown that high density of deep (and slow) interface states exist near the $\text{Al}_2\text{O}_3/(\text{Al})\text{GaN}$ interface.^{10,11} Hence, an alternative passivation method using an AlN layer has been considered for GaN-based devices.^{12–14} It was demonstrated that the plasma enhanced atomic layer deposition (PEALD)-AlN as interfacial

passivation layer is beneficial to improve the quality of $\text{Al}_2\text{O}_3(\text{HfO}_2)/\text{InGaAs}$ interface, featuring good electrical characteristics and low interface trap density.^{15,16} Wu *et al.* also observed similar passivation effect in $\text{Al}_2\text{O}_3/\text{GaN}$ structure using a 2 nm thick AlN.¹⁷ Due to the easy crystallization of AlN, however, the reverse leakage current become high when employing an AlN layer in the dielectric layer.

In our previous work, we found that the tunneling barrier height in $\text{Al}_2\text{O}_3/\text{GaN}$ interface with a 1 nm thick AlN interlayer was lower than 1.0 eV, which could be due to the inferior Al_2O_3 film quality.¹⁸ We also observed in Pt/AlN/n-GaN junctions with different AlN thicknesses that the formation of AlN layer near the AlN/GaN interface was observed clearly when the AlN thickness is above 1.5 nm.¹⁹ These suggest that at least 2 nm thick AlN grown on GaN will be beneficial to passivate the GaN surface and to improve the overgrown film quality. Although molecular beam epitaxy (MBE) grown Gd_2O_3 on GaN has been investigated,^{20,21} there is little knowledge regarding the interface characteristics of ALD- Gd_2O_3 on GaN. In this work, we comparatively investigated the interfacial and electrical properties of Gd_2O_3 on GaN with an AlN layer.

2. Experimental

Hydride vapor phase epitaxy (HVPE)-grown, n-type, c -plane (0001) GaN substrate (thickness: 300 μm , carrier concentration: $2 \times 10^{17} \text{ cm}^{-3}$) purchased from Lumistal (Korea) was used in this work. After cutting the wafer into small pieces, some of them were loaded into a thermal ALD chamber (Atomic classic, CN-1, Korea) after cleaning process in a $\text{HCl} : \text{H}_2\text{O}$ (1 : 1) solution, and three different methods to

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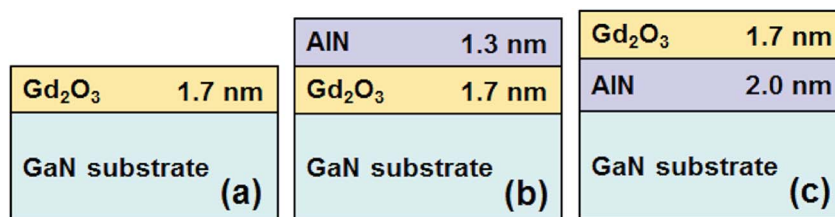


Fig. 1 Schematic layer structures of three samples denoted as (a) Gd₂O₃, (b) AlN/Gd₂O₃ and (c) Gd₂O₃/AlN.

deposit dielectric layers were applied on the prepared GaN substrates. (i) 1.7 nm thick Gd₂O₃ layer was directly deposited on the GaN surface at 250 °C with (RCp)₂Gd(L) (Air Liquide, USA) as the Gd precursor and H₂O as the oxidant (denoted as Gd₂O₃). (ii) After depositing 1.7 nm thick Gd₂O₃ on the GaN surface using the same method in (i), the temperature was ramped up to 350 °C to deposit 1.3 nm thick AlN layer using trimethylaluminum (TMA) and NH₃ as precursors (denoted as AlN/Gd₂O₃). (iii) After depositing 2 nm thick AlN on the GaN surface using the same method in (ii), 1.7 nm thick Gd₂O₃ was deposited using the same method in (i) (denoted as Gd₂O₃/AlN). The schematic layer structures of three samples are presented in Fig. 1. The thicknesses of the films were measured using a FS-1 multi-wavelength ellipsometer (Film Sense, USA). Depth resolved X-ray photoelectron spectroscopy (XPS) measurements (PHI 5000 VersaProbe, ULVAC PHI, Japan) including sputter etch treatment were carried out using a monochromatic Al K α X-ray source (beam energy: 1486.6 eV, spot size: 100 \times 100 μ m²) to observe the chemical reaction near the dielectric/GaN interface. Due to charging effects that result in a shift in binding energy, the binding energy scale was calibrated by using the adventitious C 1s peak to 284.6 eV.

The collected XPS data were analyzed using Multipak software. To characterize the electrical properties, metal–insulator–semiconductor (MIS) diodes were fabricated with a Pt Schottky electrode (diameter: 500 μ m, thickness: 50 nm) and an Al back contact (thickness: 100 nm). Capacitance–voltage (*C*–*V*) and current–voltage (*I*–*V*) measurements were performed using a HP 4284A LCR meter and Keithley 238 current source, respectively. During the *C*–*V* measurements, an AC voltage of 30 mV with various frequencies was applied to the device so that small signal conditions were realized.

3. Results and discussion

After selecting the narrow scanned XPS spectra at one etch depth (thickness of the remained AlN: \sim 1.0 nm), we compared them each other to characterize the chemical reaction near the GaN surface. Fig. 2(a) shows the Ga 2p_{3/2} core-level spectra. The peaks at \sim 1117.1, \sim 1117.4, and \sim 1118.1 eV are associated with Ga₂O, ²² Ga₂O₃ ²³ and GaN, ²⁴ respectively. The integrated intensity ratios of Ga–O/Ga–N were calculated to be 1.98, 0.52, and 0.38, respectively, for the samples with Gd₂O₃, AlN/Gd₂O₃ and Gd₂O₃/AlN layers. This indicates that the suppression of Ga–O

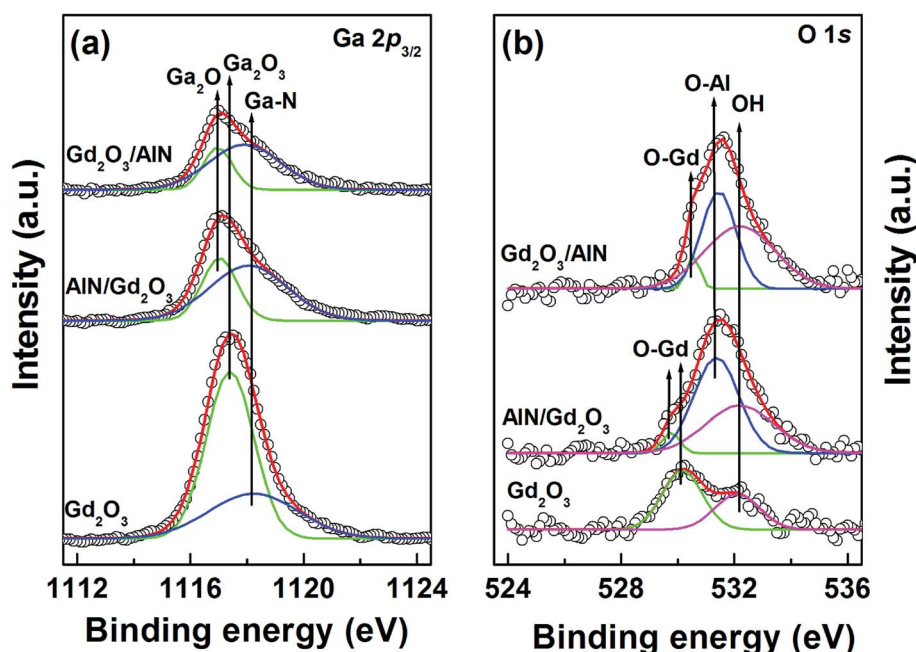


Fig. 2 XPS core level spectra of (a) Ga 2p_{3/2} and (b) O 1s for the samples with Gd₂O₃, AlN/Gd₂O₃ and Gd₂O₃/AlN layers after sputter etching. The thick of remained layer was about 1 nm.



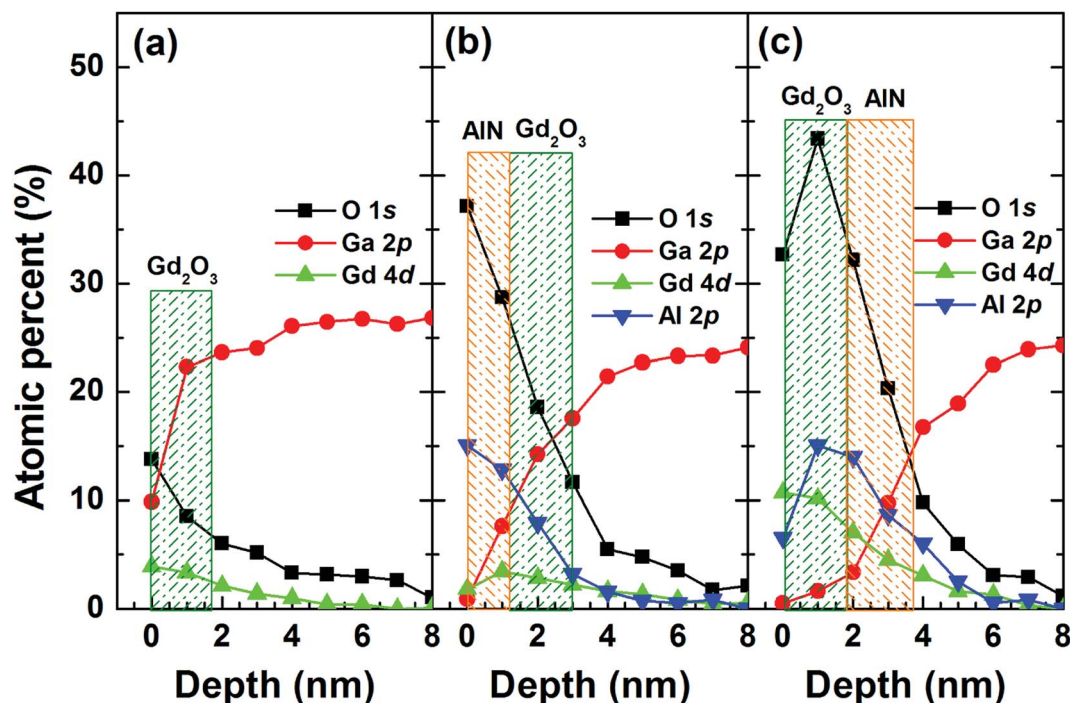


Fig. 3 XPS depth profiles for each element for the samples with (a) Gd_2O_3 , (b) $\text{AlN}/\text{Gd}_2\text{O}_3$ and (c) $\text{Gd}_2\text{O}_3/\text{AlN}$ layers.

bond occurred more significantly with a $\text{Gd}_2\text{O}_3/\text{AlN}$ layer. Not Ga_2O_3 but Ga_2O was mainly observed for the samples with $\text{AlN}/\text{Gd}_2\text{O}_3$ and $\text{Gd}_2\text{O}_3/\text{AlN}$ layers, indicating that Ga_2O_3 transferred into Ga_2O with thicker layers (so called, cleaning effect). It was shown in (In)GaAs based devices that a stable suboxide Ga_2O bond at the interface were not detrimental to device characteristics and the removal of Ga_2O_3 resulted in the reduction of frequency dispersion in capacitance.²⁵ As shown in Fig. 2(b), all three samples revealed a peak at ~ 532.5 eV, corresponding to O in the H_2O , OH^- impurities²⁶ and Gd hydroxides.²⁷ The emission peak at $529.8\text{--}530.2$ eV could be associated with O in Gd_2O_3 ,²⁷ which became weak for the samples with $\text{AlN}/\text{Gd}_2\text{O}_3$ and $\text{Gd}_2\text{O}_3/\text{AlN}$ layers. The integrated intensity ratios of $[\text{OH} + \text{O-Al}]/\text{O}_{\text{Total}}$ were found to be 0.40, 0.95, and 0.95, respectively, for the samples with Gd_2O_3 , $\text{AlN}/\text{Gd}_2\text{O}_3$ and $\text{Gd}_2\text{O}_3/\text{AlN}$ layers. It was shown that Al-OH defects commonly observed in ALD- Al_2O_3 films is the origin of acceptor-like border traps and positive fixed charges in Al_2O_3 .²⁸

Fig. 3 shows the XPS depth profiles for each element obtained from the samples. The diffusion of Ga atoms into the overgrown layer was significant. The amount of oxygen atoms near the GaN surface is similar. However, both the samples with $\text{AlN}/\text{Gd}_2\text{O}_3$ and $\text{Gd}_2\text{O}_3/\text{AlN}$ layers showed the larger amount of oxygen atoms across the overgrown layer as compared to the sample with Gd_2O_3 layer. This indicates that a significant portion of it resulted from the ALD deposition process itself. Fig. 4 shows the narrow scanned core level spectra from the sample surfaces (*i.e.*, without sputter etching). As shown in Fig. 4(a), the OH related peak observed for the samples with Gd_2O_3 and $\text{AlN}/\text{Gd}_2\text{O}_3$ layers is not observed for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer. Along with the relatively weak peak at

~ 530.2 eV corresponding to O in Gd_2O_3 , the additional peak at ~ 528.9 eV also appeared for the samples with Gd_2O_3 and $\text{Gd}_2\text{O}_3/\text{AlN}$ layers, which could be associated with more oxidized Gd-O due to excess O and lattice defects.²⁹ Fig. 4(b) shows the Gd 4d core level spectra, which mainly originated from multiplet splitting of the 4d hole with 4f valence electrons to form 9D and 7D final ionic state.³⁰ As shown in Fig. 4(c), the peak related with AlN at $73.6\text{--}73.9$ eV³¹ is dominant for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer whereas the peak associated with Al-O at ~ 74.6 eV³¹ is dominant for the sample with an $\text{AlN}/\text{Gd}_2\text{O}_3$ bilayer. The additional peak at ~ 75.2 eV with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer can be conformed to the Al-O chemical bond of Al_2O_3 or $\text{Al}(\text{OH})_3$.³²

Fig. 5(a)–(c) show the C - V curves measured at 1 kHz and 1 MHz. The frequency dispersion is hardly visible in the deep depletion region and small frequency dispersion is observed in the depletion, indicating the presence of interface states. The capacitance in accumulation at 1 kHz is much higher than that at 1 MHz, which can be due to current leakage through the dielectric layer which distorts the 1 kHz measurement.³³ The frequency dispersion in the accumulation region can also be associated with the formation of an inhomogeneous layer at the interface. Fig. 5(d)–(f) show the C - V hysteresis plots by sweeping the voltage from deep depletion to accumulation (up) and then accumulation to deep depletion (down). From the flatband capacitance (C_{FB}) obtained at each voltage sweep, the flatband voltage (V_{FB}) was calculated and then, the average interface and oxide trap density (Q_{T}) along the GaN bandgap (E_{g}) were obtained through $Q_{\text{T}} = (C_{\text{OX}}\Delta V_{\text{FB}})/qE_{\text{g}}$.³⁴ The trapped charge densities were calculated to be 3.68×10^9 , 1.82×10^{11} , and $1.83 \times 10^{12} \text{ cm}^{-2} \text{ eV}^{-1}$, respectively, for the samples with Gd_2O_3 , $\text{AlN}/$



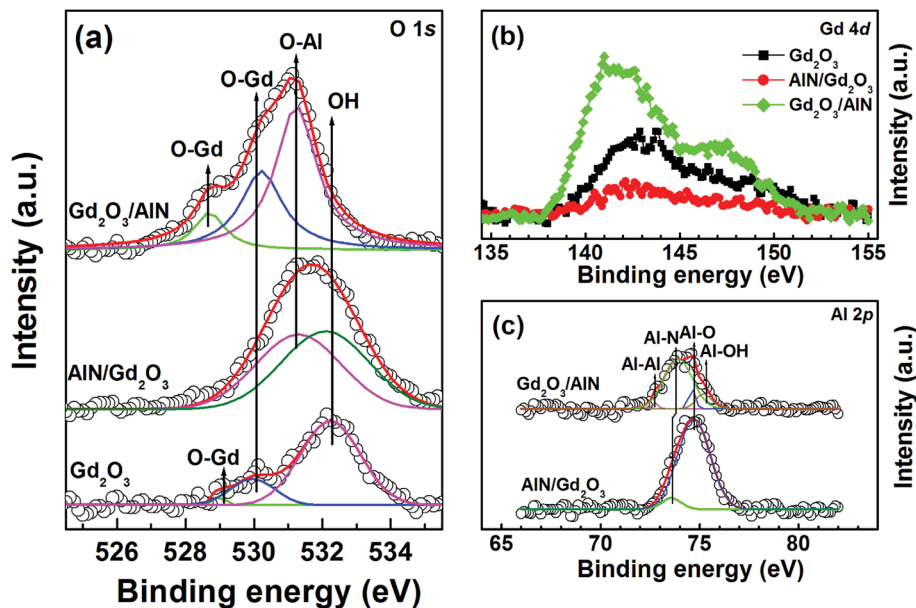


Fig. 4 XPS core level spectra of (a) O 1s, (b) Gd 4d and (c) Al 2p, which were obtained from the surface.

Gd₂O₃, and Gd₂O₃/AlN layers. The hysteresis can result from the interface states and oxide traps in the dielectric layer. Oxygen related defects are generated much with including an AlN layer (for example, see Fig. 2(b)). High density of Al–OH defects acting as acceptor-like border traps and positive fixed charges in Al₂O₃ may also increase the V_{FB} shift. Fig. 5(f) also shows that the C – V curves reveal a plateau region, reflecting the presence of 2 dimensional electron gas (2DEG) at the AlN/GaN interface.^{35,36} With increasing the bias voltage, electrons start to accumulate at the channel interface, thus forming the 2DEG. An increase in capacitance is also observed at gate bias beyond +5 V. This is due to a charge spillover from the 2DEG channel, which brings

some of the carriers closer to the surface, decreasing the effective barrier layer thickness and increasing the capacitance.³⁷

Frequency dependent conductance–voltage measurements were performed to differentiate the types of the traps. The parallel conductance (G_p/ω) can be derived by $G_p/\omega = [\omega G_m(C_{OX})^2]/[(G_m)^2 + \omega^2(C_{OX} - C_m)^2]$, where C_m is the measured capacitance, G_m is the measured conductance, and C_{OX} is the oxide capacitance. The G_p/ω values as a function of radial frequency ($\omega = 2\pi f$) can be correlated with the trap density D_T and trap response time τ_T , given by³⁸

$$\frac{G_p}{\omega} = \frac{qD_T}{2\omega\tau_T} \ln[1 + (\omega\tau_T)^2] \quad (1)$$

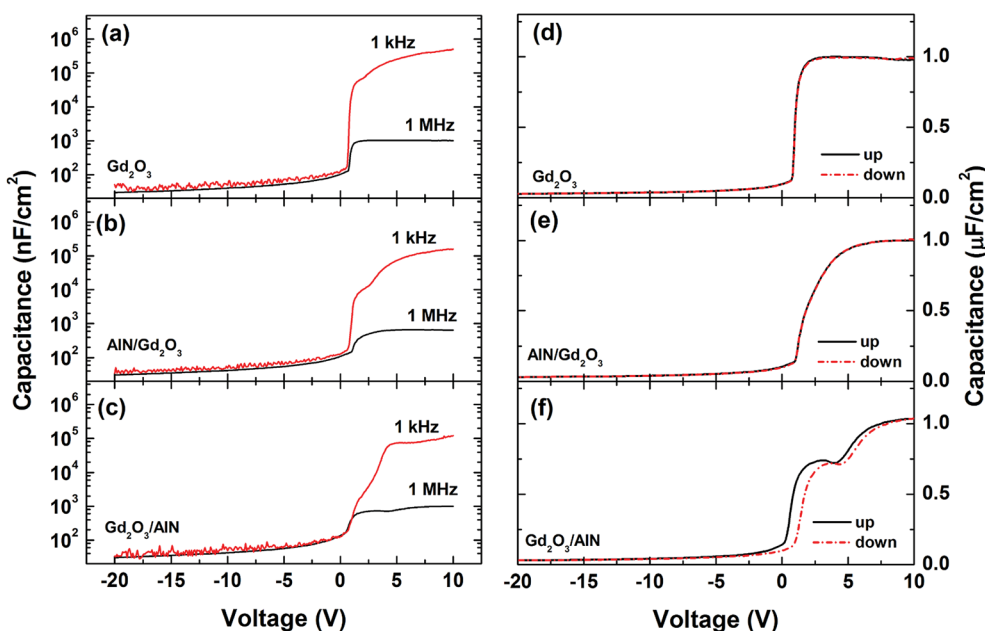


Fig. 5 (a)–(c) Capacitance–voltage (C – V) data measured at 1 kHz and 1 MHz and (d)–(f) C – V hysteresis plots measured at 1 MHz.



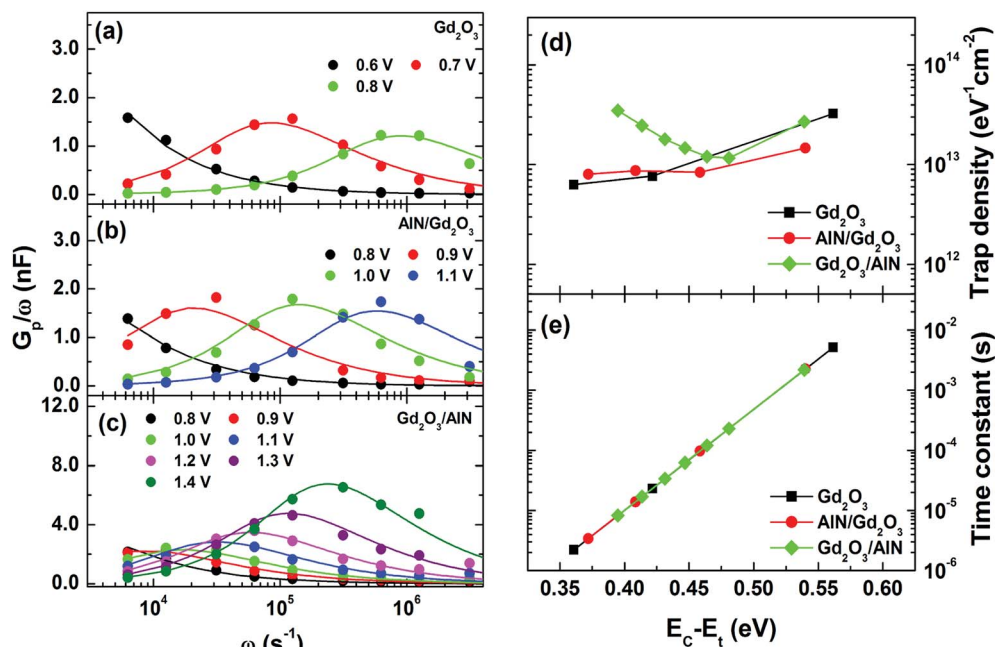


Fig. 6 Frequency dependent parallel conductance for the samples with (a) Gd_2O_3 , (b) $\text{AlN}/\text{Gd}_2\text{O}_3$ and (c) $\text{Gd}_2\text{O}_3/\text{AlN}$, and (d) the obtained trap density and (e) the time constant from the conductance method.

The experimental and fitting data are shown in Fig. 6(a)–(c). The energy level of traps below the GaN conduction band, denoted as $E_C - E_t$ (location of the trap energy) can be determined from τ_T according to the Shockley–Read–Hall statistics^{39,40}

$$\tau_T = \frac{1}{v_{th}\sigma_n N_C} \exp\left(\frac{E_C - E_t}{kT}\right) \quad (2)$$

where v_{th} is the thermal velocity, σ_n is the electron capture cross section, and N_C is the effective density of states in the GaN conduction band. Using the values presented in ref. 40, the trap density and time constant as a function of energy level were calculated, which are shown in Fig. 6(d) and (e), respectively. According to the calculated time constants, the samples with Gd_2O_3 and $\text{AlN}/\text{Gd}_2\text{O}_3$ layers showed the wider distributions compared to the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer. Interestingly, the trap density at $E_C - E_t = 0.38$ – 0.47 eV showed the highest value for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer. Hence, these traps can be considered to be from the AlN layer in the $\text{Gd}_2\text{O}_3/\text{AlN}/\text{GaN}$ structure.

As shown in Fig. 7(a), an estimate of the interface state density (D_{it}) was made by applying Terman method to the experimental C – V curves measured at 1 MHz.³⁸ The obtained D_{it} vs. $E_C - E_t$ plots in Fig. 7(b) revealed the lowest interface state density for the sample with a Gd_2O_3 layer. Interestingly, the samples with $\text{AlN}/\text{Gd}_2\text{O}_3$ and $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayers showed the highest interface state densities in the energy ranges of 0.1–0.2 eV and 0.4–0.6 eV, respectively. As shown in Fig. 3, large amount of Al atoms are present near the $\text{Gd}_2\text{O}_3/\text{GaN}$ interface for the sample with an $\text{AlN}/\text{Gd}_2\text{O}_3$ bilayer. This might lead to the formation of AlN, leaving many nitrogen vacancies at the GaN surface. Fang *et al.* reported that several kinds of deep levels can be increased at the GaN surface after Cl_2 -based inductively

coupled plasma (ICP) etching.⁴¹ In particular, they attributed a deep center located at $E_C - 0.17$ eV to nitrogen vacancy-related defect due to ICP etching. Yatabe *et al.* also showed in $\text{Al}_2\text{O}_3/\text{AlGaN}/\text{GaN}$ structure that the C – V curves in the depletion region are stretched out more significantly to the positive direction when the interface state density peaked at $E_C - 0.1$ eV is higher.⁴² Significant stretch out to the positive direction and high interface state density in the range of 0.1–0.2 eV observed for the sample with an $\text{AlN}/\text{Gd}_2\text{O}_3$ bilayer, thus, can be associated with the nitrogen vacancy-related defects. Meanwhile, Freedman *et al.* showed in $\text{AlN}/\text{AlGaN}/\text{GaN}$ MIS diodes that the trap density due to the AlN and AlGaN layers were located at 0.4–0.52 and 0.32–0.34 eV, respectively.⁴³ The energy distribution corresponding to the AlN insulating layer is similar to our work at the $\text{Gd}_2\text{O}_3/\text{AlN}$ interface. Furthermore, Fig. 7(a) shows that flatband voltage shifted negatively for the sample with a Gd_2O_3 layer as compared to the sample with an $\text{AlN}/\text{Gd}_2\text{O}_3$ bilayer. It was shown that when Gd can only exist in the +3 oxidation state, the Gd_2O_3 cannot supply oxygen effectively to the dielectric/Si interface, which produced oxygen vacancies causing the negative shift in flatband voltage.⁴⁴ As shown in Fig. 2(b), the strong Gd_2O_3 related emission observed for the sample with a Gd_2O_3 layer was not observed for the sample with an $\text{AlN}/\text{Gd}_2\text{O}_3$ bilayer. Instead, the emission related with more oxidized Gd–O bond was observed. Therefore, it can be expected that oxygen vacancies have been reduced after depositing AlN layer with the excess O atoms.

Fig. 8(a) shows the typical semi-logarithmic current density–voltage (J – V) curves measured at room temperature. Compared to the sample with a Gd_2O_3 layer, the reverse leakage current increased for the samples with $\text{AlN}/\text{Gd}_2\text{O}_3$ and $\text{Gd}_2\text{O}_3/\text{AlN}$ layers. Forward current at high bias region increased most



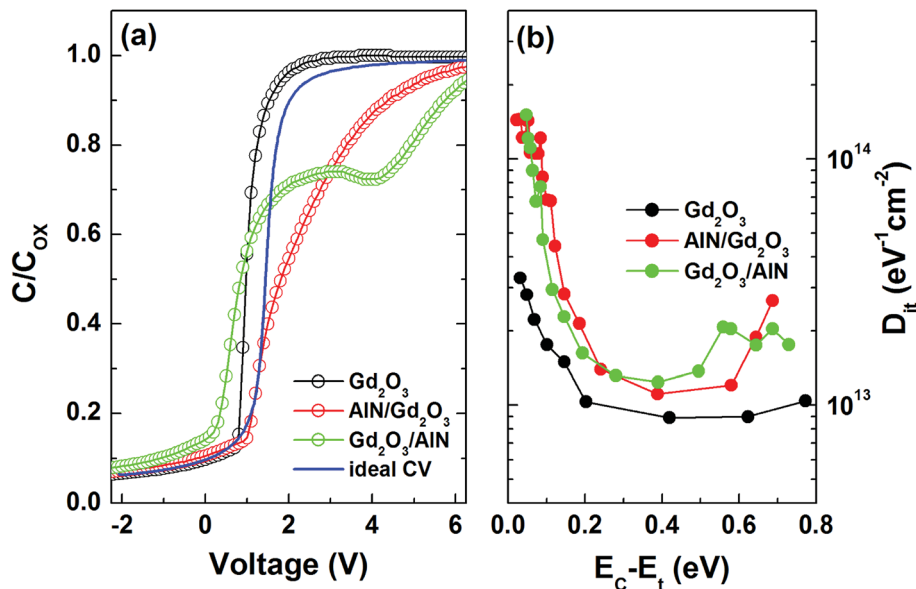


Fig. 7 (a) Comparison of experimental capacitance–voltage (C – V) data measured at 1 MHz and ideal C – V data and (b) interface state density (D_{it}) distributions determined by applying Terman method.

slowly for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer, indicating the high series resistance. The reverse leakage current density at high bias region were analyzed using the Fowler–Nordheim (FN) tunneling model, given by⁴⁵

$$J \propto E^2 \exp(-\beta/E) \quad (3)$$

where $\beta = 6.83 \times 10^{-7}(m^*)^{1/2}(\Phi_B)^{3/2}$, m^* is the effective electron mass in the insulator and Φ_B is the tunneling barrier height. As shown in Fig. 8(b), FN emissions were observed for the samples

with Gd_2O_3 and $\text{AlN}/\text{Gd}_2\text{O}_3$ layers. The tunneling barrier heights (*i.e.*, conduction band offset at $\text{Gd}_2\text{O}_3/\text{GaN}$) were calculated to be about 1.95 and 1.36 eV, respectively, for the samples with Gd_2O_3 and $\text{AlN}/\text{Gd}_2\text{O}_3$ layers. Considering the reported value of 1.9 eV in $\text{Gd}_2\text{O}_3/\text{GaN}$ interface,⁴⁶ the value for the sample with a Gd_2O_3 layer is very similar. Relatively lower value for the sample with an $\text{AlN}/\text{Gd}_2\text{O}_3$ bilayer may be due to the poorer interfacial quality as well as the interface and oxide traps.

Fig. 8(b) also shows that no FN emission was observed for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer. Instead of FN emission,

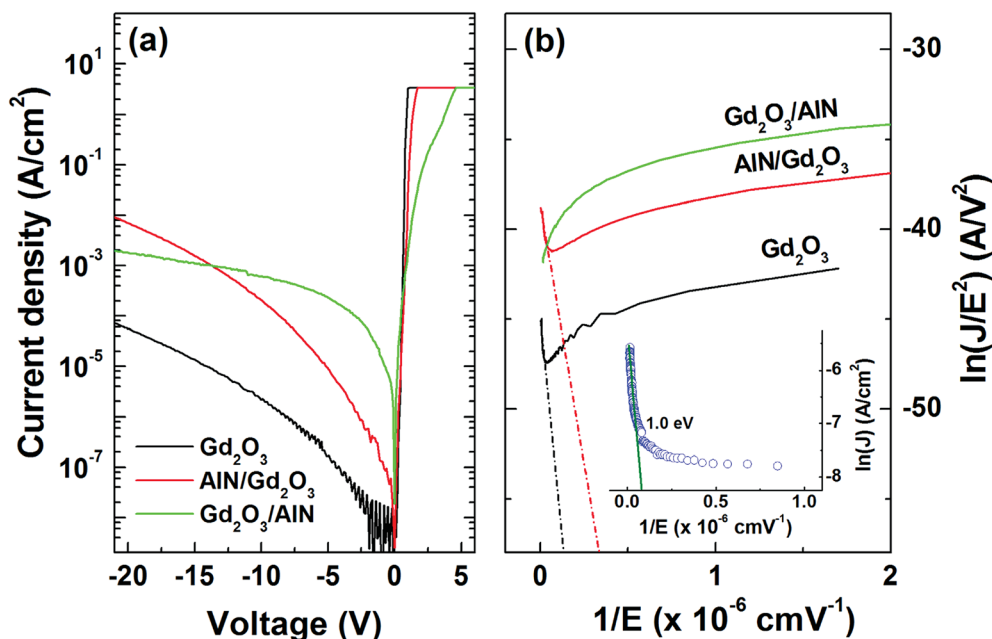


Fig. 8 (a) Typical semi-logarithmic current density–voltage (J – V) characteristics and (b) Fowler–Nordheim (FN) tunneling plots of $\ln(J/E^2)$ vs. $1/E$. The inset in (b) presents trap assisted tunneling (TAT) plot of $\ln(J)$ vs. $1/E$.



the trap assisted tunneling (TAT) was applied to the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer. In TAT emission, the reverse leakage current is given as $J \propto \exp(-\gamma/E)$, where $\gamma = 6.83 \times 10^{-7}(m^*)^{1/2}(\Phi_t)^{3/2}$ and Φ_t is the trap energy level, which can be obtained from the $\ln(J)$ vs. $1/E$ plot. From the linear fittings to the data as shown in the inset in Fig. 8(b), the trap energy level was estimated to be 1.00 eV. Energetically most favorable point defects in AlN are substitutional oxygen for nitrogen (O_N) and aluminum vacancy (V_Al), with the energy levels of ~ 0.8 and ~ 1.0 eV, respectively.⁴⁷ It was shown that dangling bonds in Al give rise to ~ 1.0 eV in energy level.⁴⁸ At present moment, it is suggested that Al related defects are responsible for TAT emission for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer.

4. Conclusions

In conclusion, we investigated the interfacial and electrical properties of atomic layer deposited Gd_2O_3 with an AlN layer on n-GaN. X-ray photoelectron spectroscopy (XPS) spectra showed the dominant peaks associated with Ga–O and O–Al bonds, respectively, near the $\text{Gd}_2\text{O}_3/\text{GaN}$ and $\text{AlN}/\text{Gd}_2\text{O}_3/\text{GaN}$ and $\text{Gd}_2\text{O}_3/\text{AlN}/\text{GaN}$ interfaces. Large amount of oxygen atoms across the dielectric layers were observed for the samples with $\text{Gd}_2\text{O}_3/\text{AlN}$ and $\text{AlN}/\text{Gd}_2\text{O}_3$ layers. The highest flatband voltage shift in C – V hysteresis characteristics were observed for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer, indicating the highest interface and oxide trap densities. According to parallel conductance and Terman methods, the samples with $\text{AlN}/\text{Gd}_2\text{O}_3$ and $\text{Gd}_2\text{O}_3/\text{AlN}$ layers were found to have the highest interface state densities in the energy ranges of 0.1–0.2 eV and 0.4–0.6 eV, respectively. The reverse leakage currents for the samples with Gd_2O_3 and $\text{AlN}/\text{Gd}_2\text{O}_3$ layers were explained by Fowler–Nordheim (FN) whereas trap assisted tunneling (TAT) was more appropriate for the sample with a $\text{Gd}_2\text{O}_3/\text{AlN}$ bilayer.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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