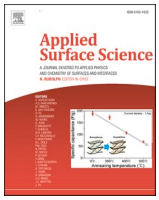
Applied Surface Science 622 (2023) 156954

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc



Short Communication

Interface dipole induced threshold voltage shift in the Al_2O_3/GaN heterostructure

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A B S T R A C T

The Al_2O_3/GaN heterostructure is a crucial component of GaN-based electronic and photonic devices, and the Al_2O_3/GaN interface quality plays an important role in determining the device performance. Here, using density functional theory, we confirmed that dipole formed at the Al_2O_3/GaN interface can be attributed to electron transfer and redistribution between Al_2O_3 and GaN. The formation of dipole was confirmed by X-ray photoemission spectroscopy. The dipole induced electric field at the interfaces of the Al_2O_3/GaN heterostructures result in negative threshold voltage (V_{TH}) shifts of 2.3 and 1.2 V for the heterostructures without defects and with one Al interstitial (Al_i) defect, respectively. On the other hand, the Al_i defect can induce positive charges, resulting in negative V_{TH} shifts. Therefore, an improvement in the interface quality (i.e., by eliminating the Al_i defect) does not necessarily result in positive V_{TH} shifts. This study reveals novel electronic properties of the Al_2O_3/GaN heterostructure and offers a path toward the achievement of GaN-based devices with engineered features.

1. Introduction

GaN-based, high electron mobility transistors (HEMTs) can deliver a high current, a high voltage, and high switching speeds. As such, they are widely used in high power and high radio frequency devices [1–3]. Gate dielectrics are widely used to suppress the gate leakage current and enlarge the gate breakdown voltage in GaN HEMTs [4,5]. The Al_2O_3 fabricated by atomic layer deposition (ALD) has been broadly adopted as a gate dielectric material owing to its superior electronic properties, including its high bandgap and dielectric constant [6–8]. However, although the Al_2O_3 obtained via ALD exhibits superior properties, numerous interface positive fixed charges and traps are introduced at the Al_2O_3/GaN interface or in the bulk Al_2O_3 [8,9]. These positive, fixed charges can induce a negative threshold voltage (V_{TH}) shifts, hindering the development of enhancement-mode GaN HEMTs. Moreover, traps at the Al_2O_3/GaN interface can result in reliability and stability challenges. [7,9–11] The occurrence of positive charges and traps at the Al_2O_3/GaN interface can be attributed to the defects at the interface. Non-stoichiometric atomic ratios of O and Al have been found at the Al_2O_3/GaN interface, with Al significantly surpassing O [12,13]. The Al-rich conditions can produce excess Al interstitial (Al_i) defects. Generally, structural impurities, such as Al_i defects and O vacancies, can introduce various defect levels in the Al_2O_3 bandgap [14,15]. The charged defect levels can behave like fixed charges or fast and slow traps depending on their energy levels in the bandgap [14,15]. When the energy states are above the GaN conduction band edges, under either positive or negative gate stress on the Al_2O_3/GaN heterostructure, the Fermi level can hardly modulate these energy levels Thus, the defects behave similarly to fixed charges [10,14,15]. However, when the energy states are within the bandgap of GaN and close to the midgap, charge trapping and detrapping occurs when the Fermi level moves across the energy levels. Resultantly, the defects behave similarly to trap states [14,15]. Considerable efforts have been devoted to improving the interface quality between gate dielectrics and GaN [7,9,11]. Although the interface trap density can be reduced to an extremely low value, in many cases, the V_{TH} does not shift to a positive value accordingly or it may even shift to a more negative value [9,16]. As both the fixed charges and interface traps are ascribed to the interface defects [14,15], the positive charges thus the positive charges induced negative V_{TH} shifts should also be suppressed together with the interface trap density. The abnormal V_{TH} shifts after the improvement in the interface quality indicates that there are additional factors that contribute to the V_{TH}, which were not considered prior.

In addition to the interface charges and traps, interface dipoles control the interface electronic structures [17–23]. Generally, dipoles are formed between dielectrics and semiconductors or between two dielectrics, and they have a significant impact on the device performance. For example, dipoles formed at the HfO_2/Si interface have been found to negatively shift V_{TH} [17–19]. The origins of the dipoles are unclear; however, the formation of the Si–O–Hf bonds could be one

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https://doi.org/10.1016/j.apsusc.2023.156954
Received 2 January 2023; Received in revised form 18 February 2023; Accepted 3 March 2023
Available online 9 March 2023

reason for the dipole formation [17,19]. Dipoles were also widely found at high-k/SiO_2 interface, and the magnitude of the V_{TH} shifts relies on the employed high-k materials [20–22]. The electronegativity difference between the cations of high-k and SiO_2 is regarded as one reason for the dipole formation [22]. Another possible reason is the oxygen density difference at the junctions between the high-k materials and SiO_2 [20]. Moreover, dipole formation could occur between two high-k materials, such as HfO_2 and Al_2O_3, and dipoles at the HfO_2/Al_2O_3 interface have been applied to explain the abrupt V_{TH} shifts [23]. Based on these results, we hypothesize that dipoles can be formed at the Al_2O_3/GaN interface. In addition to the defect induced charges, dipoles could be an additional factor affecting the V_{TH} of the Al_2O_3/GaN heterostructure.

2. Experiments

We perform spin-polarized first principles calculations within the framework DFT, as implemented in the Vienna ab-initio simulation package. The total energy convergence threshold is set to 10^{-6} eV and the structures are optimized until the Hellmann-Feynman forces stay below 0.01 eV/Å. All the calculations are performed within the Perdew-Burke-Ernzerhof generalized gradient approximation. An energy cutoff of 450 eV is used in the plane wave expansion. The θ-phase Al_2O_3 was adopted to build the Al_2O_3/GaN heterostructure, as its bandgap and mass density values are similar to those of amorphous Al_2O_3 grown by ALD [8]. The bottom dangling bonds of GaN in the Al_2O_3/GaN heterostructures were passivated by pseudohydrogen with 0.75 valence electrons to ensure that the density of states in the bandgap are from the interface defects [24]. In the subsequent DFT calculations, one Al_i defect was introduced into the Al_2O_3/GaN interface to evaluate its effect on the electronic properties. For the X-ray photoemission spectroscopy (XPS) measurements, 1.2 nm Al_2O_3 was deposited on the surface GaN (0001). Before the ALD Al_2O_3 film growing process, the GaN substrates were cleaned with diluted hydrochloric acid for 3 min to remove the surface native oxide layers. Subsequently, the GaN substrates were transferred to the ALD chamber for Al_2O_3 deposition. One ALD Al_2O_3 cycle comprised a 0.015 s trimethyl aluminum dose pulse and a 4 s O_2 plasma oxidization, followed by a 3 s purge with N_2. The ALD Al_2O_3 growth per cycle was 1.2 Å, measured by an ellipsometer.

3. Results and discussion

The insets of Fig. 1(a) and (b) show the atomic structures of the Al_2O_3/GaN interface regions without and with one Al_i defect, respectively. The Al_i defect is indicated by an arrow in Fig. 1 (b). After introducing one Al_i defect, the atomic positions of Al, Ga, and O were severely distorted at the interface compared with the case of the heterostructure without defects. For the Al_2O_3/GaN heterostructure without the Al_i defect, eight Ga–O bonds were initially observed at the interface, with the eight oxygen atoms perfectly saturating the eight Ga dangling bonds [8]. The N-Ga-O-Al interface bonding used in this study is the most stable interface configuration of the Al_2O_3/GaN heterostructure [25]. Fig. 1(a) shows the total density of states (TDOS) without the Al_i defect. The electron counting rule was satisfied in the Al_2O_3/GaN heterostructure, thus leading to an insulating interface with a clean bandgap. The strong peak located at 3.2 eV can be attributed to contribution from the Al atoms (Fig. S1 and 2). However, as shown in Fig. 1(b), gap states occur after the introduction of the Al_i defect. In addition, the Al_2O_3/GaN heterostructure with the Al_i defect has a large TDOS close to 3.2 eV, therefore, the peak located at 3.2 eV of the heterostructure without the Al_i defect is not observable. As discussed above, the interface states occur in the bandgap can behave as traps or fixed charges, thereby deteriorating the device performance.

Charge density difference was calculated to analyze the role of Al_i defects on the charge transfer and redistribution at the Al_2O_3/GaN interfaces. The electron densities of Al_2O_3 and GaN were subtracted from those of the Al_2O_3/GaN heterostructures. Fig. 2(a) and (b) show the Al_2O_3/GaN heterostructures without and with the Al_i defect, respectively. The charge transfer and redistribution primarily occur at the N–Ga–O–Al layer, with negligible change on charge density in the inner Al_2O_3 and GaN far away from the interfaces. The charge density difference at the Al_2O_3/GaN interfaces indicates electron transfer from GaN to Al_2O_3 across the interfaces. Charge accumulation (yellow part) primarily occurs around the O layer in Al_2O_3, with a small accumulation area observed around the N layer in GaN; furthermore, strong charge depletion (light blue part) occurs around the Ga layer in GaN, with a small depletion area observed around the Al layer in Al_2O_3. Consequently, in the equilibrium state, dipoles form at the Al_2O_3/GaN interfaces without and with the Al_i defect as the built-in electric field is established. The built-in electric field is oriented from GaN to the Al_2O_3, which can induce a negative V_{TH} shifts. As shown in Fig. 2(b), after interposing one Al_i defect, the charge accumulation around the O layer and charge depletion around the Ga layer reduced considerably. Consequently, the dipole induced electric field could be effectively attenuated accordingly. The dipole induced V_{TH} shifts in the Al_2O_3/GaN heterostructures without and with the Al_i defect can be estimated based on the equation[23]:

$$V_{TH} = -\left(\frac{\int_0^{d_1} dz \int Q(x,y,z)dxdy + \int_0^{d_2} dz \int Q(x,y,z)dxdy}{\varepsilon_{GaN}} + \frac{\int_0^{d_3} dz \int Q(x,y,z)dxdy + \int_0^{d_4} dz \int Q(x,y,z)dxdy}{\varepsilon_{Al2O3}}\right)$$

where d_1, d_2 and d_3, d_4 shown in Fig. 2 (a) and (b) are the widths of the dipole layer across the GaN and Al_2O_3 layers, respectively. As shown in Fig. 2 (a), the values of d_1, d_2, d_3, and d_4 are 1.72, 1.62, 1.14, and 1.72 Å,

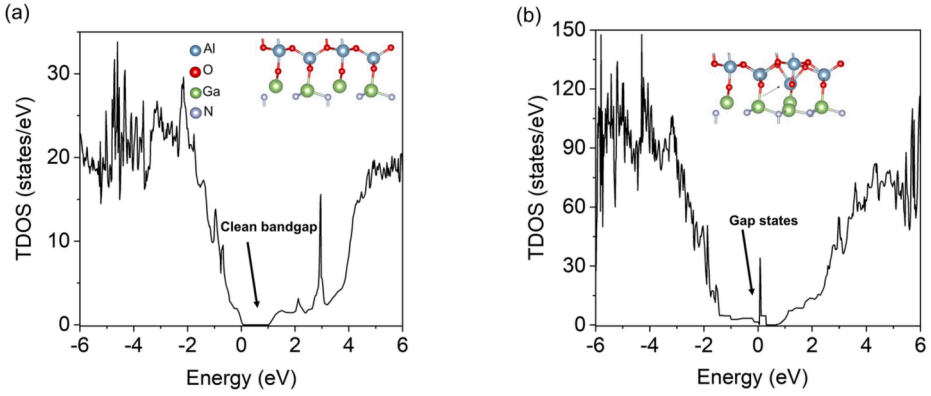

Fig. 1. (a) Total density of states of the Al_2O_3/GaN heterostructures (a) without and (b) with the Al_i defect. Inset shows the atomic structure configuration at the interfaces where the Al_i defect is indicated by an arrow in (b).

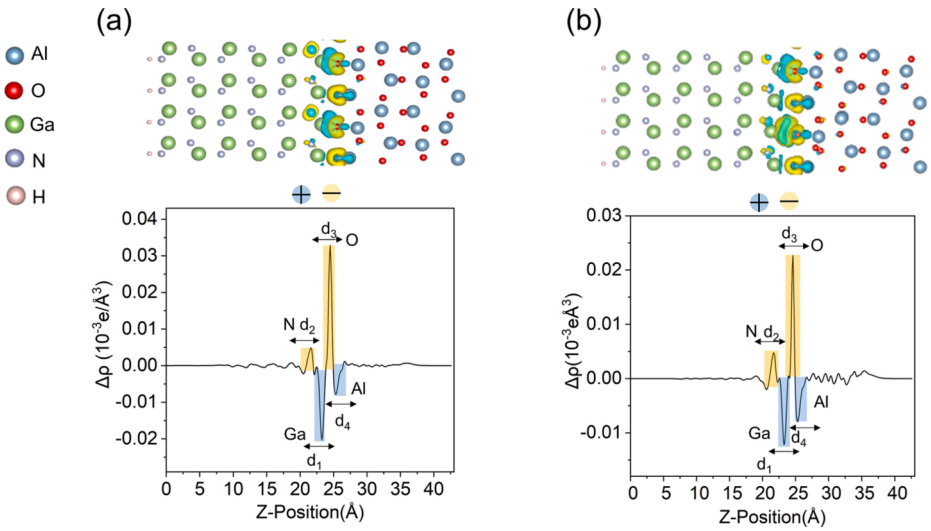

Fig. 2. Charge density differences of Al_2O_3/GaN heterostructures (a) without and (b) with the Al_i defect and the corresponding planar-averaged, electron density difference of the heterostructures.

respectively. As shown in Fig. 2 (b), the values of d_1, d_2, d_3, and d_4 are 1.72, 1.52, 0.95, and 1.62 Å, respectively. Q(r) is the volume charge density; ε_{GaN} and ε_{Al2O3} are the dielectric constants of GaN and Al_2O_3, respectively. According to the equation, the negative V_{TH} shifts of 2.3 V can be reduced to 1.2 V after interposing one Al_i defect. The DFT results here imply that even without any interface defects, owning to the electron transfer and redistribution at the Al_2O_3/GaN interfaces, the dipole induced electric field can still induce a negative V_{TH} shifts. Therefore, the dipole formed at the interface is the intrinsic property of the Al_2O_3/GaN heterostructure, and thus, it cannot be eliminated by removing the interface defects. In addition, the above DFT calculation was based on the Ga-polar GaN. However, for the N-polar GaN, charge transfer will also across the N-Ga-N-Al interface chemical bonds where the interface dipole forms at the interface of the Al_2O_3/GaN (N-polar) heterostructure.

The Al_i defect in Al_2O_3 can introduce positive charges and induce a negative V_{TH} shifts [14,15,26]. Therefore, after eliminating the Al_i defect, the defect induced negative V_{TH} shifts will be suppressed. Substantial efforts have been devoted to improving the interface quality between Al_2O_3 and GaN [7,9,11]. Although the interface trap density can be reduced to an extremely small value, the V_{TH} does not change correspondingly or it may even exhibit a negative shift [9,16]. As both the fixed charges and interface traps are ascribed to the defects at the interface [14,15], the positive charges and the positive charges induced negative V_{TH} shifts should be suppressed together with the interface trap density. In this study, the abnormal V_{TH} movement after interface quality improving of the Al_2O_3/GaN heterostructures is attributed to the dipole induced negative V_{TH} shifts can be increased from 1.2 to 2.3 V after removing the Al_i defect at the interface. Except from positive charge induced negative V_{TH} shifts, dipole is an additional factor that can modulate the V_{TH} in the Al_2O_3/GaN heterostructure. Fig. 3(a) shows the Ga 3*d* XPS spectra of the Al_2O_3/GaN heterostructure and pristine GaN. The XPS peak located at 19.7 eV corresponds to Ga-N bond in the pristine GaN, while the Ga-O bond located at 20.7–20.9 eV is absent in the Al_2O_3/GaN heterostructure [27]. Therefore, no oxidization occurs during the ALD process. Compared with that for the pristine GaN, after the heterostructure formation, the binding energy of Ga 3*d* shifts toward a lower value (~0.5 eV). Additionally, N 1 *s* XPS spectra also shifts to a smaller value after Al_2O_3 coating (Fig. S3). Based on the above DFT calculation results, it was established that electron redistribution mainly occurred at the Al_2O_3/GaN interface, and the dipole induced electric field is oriented from GaN to Al_2O_3. Fig. 3(b) shows the band alignment diagram of the Al_2O_3/GaN heterostructure before and after Al_2O_3 coating. Owning to the dipole induced electric field, upward band bending occurs at the interface of the Al_2O_3/GaN heterostructure (0.48 eV), which can shift the binding energy of Ga 3*d* toward a lower value [28]. The value of band bending was obtained from the valence band spectra (Fig. S4). Fig. 4(a) and (b) display 2D electron localization function (ELF) profiles of Al_2O_3/GaN heterostructures without and with the Al_i defect, respectively. The ELF value varies from 0 to 1, where the value smaller than 0.5 indicates the delocalization of the electrons and a larger value indicates the stronger ability to accumulate electrons [29].

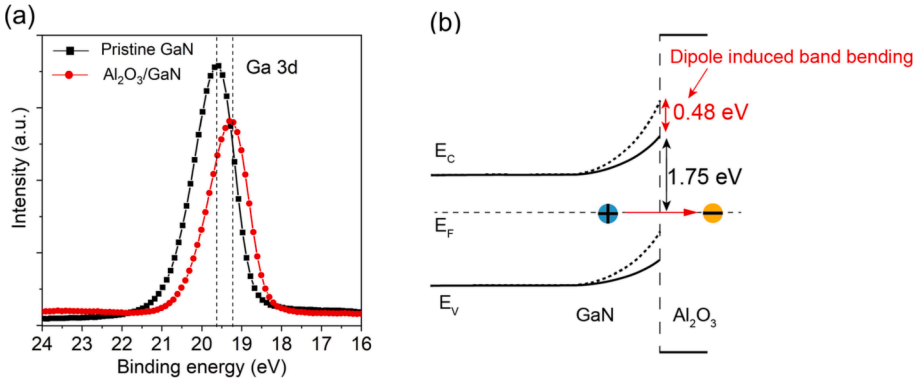

Fig. 3. (a) Ga 3*d* XPS spectra for the Al_2O_3/GaN heterostructure and pristine GaN. (b) Schematic band alignment diagram of the Al_2O_3/GaN heterostructure where the dipole induced electric field is also shown. The solid and dashed lines correspond to the band bending before and after Al_2O_3 coating.

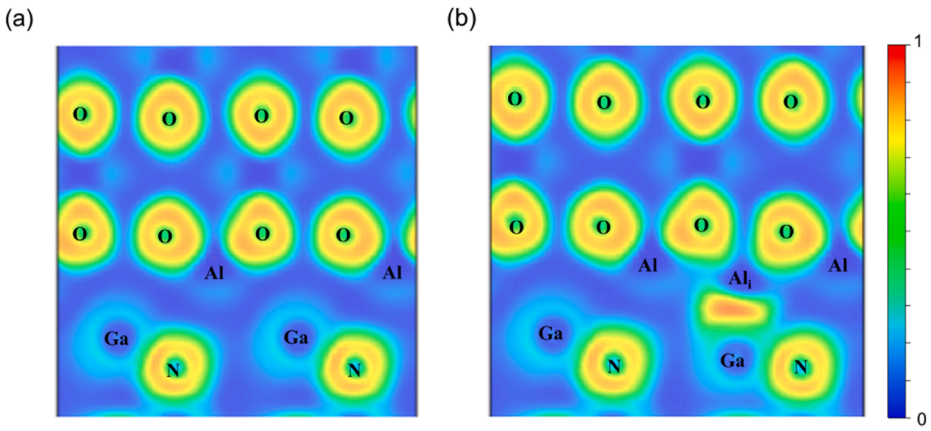

Fig. 4. 2D ELF profiles of Al_2O_3/GaN heterostructures (a) without and (b) with the Al_i defect.

The charge depletion and accumulation of each atom are labeled correspondingly in Fig. 4(a) and (b). After interposing one Al_i defect, strong electron accumulation occurs between the Ga atoms and Al_i defects, indicating the formation of Ga–Al bonds at the Al_2O_3/GaN interface. The strong electron accumulation accounts for the stability of the Al_2O_3/GaN heterostructure with the Al_i defect. The Ga–Al bond can potentially change the polarity of the GaN surface from the Ga face to the N face [10,30]. As Ga-face and N-face GaN exhibit polarization charge densities of -2×10^{13} and 2×10^{13} cm^{-2}, respectively [31], the inversion of the polarity charge could be one source of the positive charges at the Al_2O_3/GaN interface.

4. Conclusions

In summary, we confirmed that dipoles formed at the Al_2O_3/GaN interface can be attributed to the electron transfer between Al_2O_3 and GaN. The magnitude of the dipole induced V_{TH} shifts can be reduced after improving the interface quality (eliminating the Al_i defect). Moreover, we found that the Al_i defect forms a stable bond with the Ga atom (Al–Ga), indicating the stability of the Al_2O_3/GaN heterostructure with the Al_i defect. In addition, because charge transfer and redistribution are fundamental properties of the interface chemistry of two dissimilar interfaces, we propose that dipole formation could occur at other high-k material/semiconductor interfaces, such as Ga_2O_3/GaN, SiN/GaN, and Al_2O_3/Si heterostructures.

CRediT authorship contribution statement

Chuanju Wang: Conceptualization, Methodology, Validation, Investigation, Writing – original draft, Writing – review & editing. **Xiaohang Li:** Resources, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Chuanju wang reports was provided by KAUST. Chuanju wang reports a relationship with King Abdullah University of Science and Technology that includes: employment.

Data availability

No data was used for the research described in the article.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apsusc.2023.156954.

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