Atomic Layer Deposition of Ultrathin La\textsubscript{2}O\textsubscript{3}/Al\textsubscript{2}O\textsubscript{3} Nanolaminates on MoS\textsubscript{2} with Ultraviolet Ozone Treatment

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Abstract: Due to the chemically inert surface of MoS\textsubscript{2}, uniform deposition of ultrathin high-k dielectric using atomic layer deposition (ALD) is difficult. However, this is crucial for the fabrication of field-effect transistors (FETs). In this work, the atomic layer deposition growth of sub-5 nm La\textsubscript{2}O\textsubscript{3}/Al\textsubscript{2}O\textsubscript{3} nanolaminates on MoS\textsubscript{2} using different oxidants (H\textsubscript{2}O and O\textsubscript{3}) was investigated. To improve the deposition, the effects of ultraviolet ozone treatment on MoS\textsubscript{2} surface are also evaluated. It is found that the physical properties and electrical characteristics of La\textsubscript{2}O\textsubscript{3}/Al\textsubscript{2}O\textsubscript{3} nanolaminates change greatly for different oxidants and treatment processes. These changes are found to be associated with the residual of metal carbide caused by the insufficient interface reactions. Ultraviolet ozone pretreatment can substantially improve the initial growth of sub-5 nm H\textsubscript{2}O-based or O\textsubscript{3}-based La\textsubscript{2}O\textsubscript{3}/Al\textsubscript{2}O\textsubscript{3} nanolaminates, resulting in a reduction of residual metal carbide. All results indicate that O\textsubscript{3}-based La\textsubscript{2}O\textsubscript{3}/Al\textsubscript{2}O\textsubscript{3} nanolaminates on MoS\textsubscript{2} with ultraviolet ozone treatment yielded good electrical performance with low leakage current and no leakage dot, revealing a straightforward approach for realizing sub-5 nm uniform La\textsubscript{2}O\textsubscript{3}/Al\textsubscript{2}O\textsubscript{3} nanolaminates on MoS\textsubscript{2}.

Keywords: atomic layer deposition; MoS\textsubscript{2}; La\textsubscript{2}O\textsubscript{3}/Al\textsubscript{2}O\textsubscript{3}; ultraviolet ozone

1. Introduction

Silicon complementary metal-oxide-semiconductor (CMOS) devices continuing shrink in size, and keeping the generation of heat low has becoming extremely challenging [1,2]. One promising alternative approach is to use transition metal dichalcogenides (TMDs) due to their extraordinary electronic and mechanical properties [3,4]. Particularly, molybdenum disulfide (MoS\textsubscript{2}) with a natural bandgap (1.2–1.8 eV) has attracted plenty of researches for its promising application in scaled low-power field effect transistors (FETs) and flexible devices [5,6]. A crucial step in the manufacturing of FETs is the growth of ultrathin and uniform high-k gate dielectric on MoS\textsubscript{2}. The mobility of MoS\textsubscript{2} can be further improved after high-k gate films deposition through the suppression of Coulomb scattering by the dielectric mismatch effect between the MoS\textsubscript{2} and high-k dielectric [7,8]. The most controlled approach for obtaining nanoscale, high-quality growth of dielectrics is atomic layer deposition (ALD). However, atomic layer deposition of ultrathin and uniform high-k gate films on MoS\textsubscript{2} still represents one of the key challenges to be addressed due to the lack of dangling bonds or nucleation sites on the MoS\textsubscript{2} surface. The physical adsorption of precursors on the surface is considered to be a key element that enables the initial ALD reaction to take place [9]. Nevertheless, the weakly physical adsorption of precursors can be easily desorbed from the surface by the subsequent purge gas [10]. The dielectric films easy to form pinhole-like defects when it is directly deposited on MoS\textsubscript{2} due to random nucleation at defects, edges, and impurities, especially the thickness of the dielectric less than a few nanometers [11].
However, to meet the demand of ultra-scaled FETs, the thickness of gate dielectric layer needs to be extremely thin (<5 nm) for sufficient electrostatic coupling of the gate to the semiconducting channel [12]. To cope with these challenges, the pretreatment of a MoS₂ surface with oxygen plasma [13], introduction of an additional seeding layer [14], ultraviolet ozone (UV-O₃) [15], water plasma treatment [16] have been demonstrated. Growth of sub-5 nm uniform Al₂O₃ film on MoS₂ has been achieved [12]. La₂O₃ has a high dielectric constant (~26), large band gap (~5.8 eV), and the drawback of moisture absorption can be greatly improved by mixed with a less hygroscopic oxide Al₂O₃ [17,18]. It has been studied as the candidate gate dielectric in the sub-22 nm technical process node. The La₂O₃/Al₂O₃ nanolaminate processed film can provide a higher dielectric constant and better leakage current control at the same physical thickness compared to the Al₂O₃ film. However, to date, the growth of La-based binary or ternary compounds on MoS₂ has not been investigated. Therefore, in this paper, the ALD deposition of sub-5 nm La₂O₃/Al₂O₃ nanolaminates on MoS₂ is carried out and the properties of La₂O₃/Al₂O₃ nanolaminates on MoS₂ are investigated.

2. Materials and Methods

In the experiment, n-type silicon (100) wafers with a resistivity of 2–4 Ω·cm were cleaned by RCA method and a 60 s dip in diluted HF solution was used to remove the native oxide, followed by 5 min of washing with deionized water. Then, the silicon wafers were immediately transferred to an ultra-high vacuum RF magnetron sputtering system chamber and a MoS₂ target was cleaned in 10 min by pre-sputtering under the deposition conditions. Afterward, few layers MoS₂ film was directly deposited by RF magnetron sputtering system with the RF power of 50 W at 400 °C. For sulfur compensation and defects reduction, all wafers were annealed in the hydrogen sulfide at 700 °C for 60 min. After that, some wafers were treated by UV ozone ProCleaner plus system under the power of 11.04 mW·m⁻² for 5 min at the room temperature to improve the surface of MoS₂. The Raman spectra of MoS₂ before and after UV-O₃ treatment is shown in Figure 1. The two characteristics Raman modes (A₁g and E'₁g) of the MoS₂ can be observed and their positions changed negligibly before and after UV-O₃ treatment. It indicates that the treatment causes minimal structural damage in MoS₂. Moreover, the difference between these two peaks is 27.3 cm⁻¹, which indicated that the thickness of MoS₂ is between five and seven layers [19].

![Figure 1. Raman spectra of MoS₂ before and after UV-O₃ treatment.](image-url)
Then, the wafers with or without UV-O$_3$ treatment were transferred to the ALD chamber to deposited La$_2$O$_3$/Al$_2$O$_3$ nanolaminates at 260 °C. Tris (isopropylcyclopentadienyl) lanthanum (La(iPrCp)$_3$) and trimethyl-aluminum (TMA) was used as the lanthanum and aluminum precursor, respectively. H$_2$O and O$_3$ was used as the oxidant, respectively. O$_3$ was generated by the ozone generator using ultra-pure O$_2$ (99.999%).$^{10}$ deposition sequence cycles of TMA/H$_2$O/La(iPrCp)$_3$/H$_2$O and TMA/O$_3$/La(iPrCp)$_3$/O$_3$ were used to obtain H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates, respectively. Before the ALD deposition sequence, a 4 s pulse time of TMA was carried out firstly to form the physical adsorption on the surface. ~3 nm H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates were measured by Woollam M2000D spectroscopic ellipsometry. After O$_3$-based and H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates deposition process, Al electrode was fabricated by photolithography patterning to form MOS capacitors after back Al electrode was prepared by magnetron sputtering. Atomic force microscopy (AFM, Bruker Dimension Edge, Bruker Nano Inc., Billerica, WA, USA), X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, Thermo Fisher Scientific Inc., Waltham, MA, USA) were used to character the properties of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$. The standard electrical measurements were performed at room temperature using the Keithley 4200SCS characterization system (Tektronix Inc., Kent, WA, USA).

3. Results and Discussion

Figure 2 shows the AFM results of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$. It can be found that non-uniformity surface is observed for both O$_3$-based and H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$, which indicates that it is difficult to grow uniform ultrathin dielectric directly on MoS$_2$. Meanwhile, a smoother surface is obtained for O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates compared to H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$. This may be explained by that, O$_3$ has higher reactivity due to its strong oxidizing ability, it is easy to decompose to O$_2$ and monatomic O during the ALD reactions, the monatomic O radical diffusion and desorption will significantly affect the growth of the film. Using ozone as oxidant enhances the Al$_2$O$_3$ film coverage and uniformity on MoS$_2$ due to ozone facilitates initial TMA precursor nucleation on the MoS$_2$ [20], which is consistent with the AFM results. After MoS$_2$ treated with UV-O$_3$, the improvement of the uniform surface is observed for both O$_3$-based and H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates (and especially for O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates). The root mean square (RMS) value of O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates decreases from 0.381 nm to 0.150 nm, while the RMS value of H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates decreases from 0.394 nm to 0.186 nm after MoS$_2$ suffered from UV-O$_3$ treatment. Generally, the lack of reaction surface for MoS$_2$ lead to an increase of surface roughness after ALD deposition due to the buildup of precursors and reaction products randomly occurred [11]. The improvement of uniform and decrease of RMS value suggest that the initial surface nucleation of ultrathin La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ can be improved by UV-O$_3$ treatment.
To evaluate the dielectric electrical properties with nanometer resolution, conductive AFM measurements are carried out by applying a constant voltage between the Pt-Ir coated tip and sample. Figure 3 shows the current images measured by conductive AFM when applying a sample bias of 1 V. As shown in Figure 3, the density of leakage dots in the H₂O-based La₂O₃/Al₂O₃ nanolaminates is higher than that in the O₃-based La₂O₃/Al₂O₃ nanolaminates. The leakage dot is an indicator of conductive paths exist in La₂O₃/Al₂O₃ nanolaminates. They are not only attributed to surface roughness, but also possibly caused by local fluctuations in composition and/or structures, and/or by defects in La₂O₃/Al₂O₃ nanolaminates [21]. The presence of many leakage dots indicates that H₂O-based La₂O₃/Al₂O₃ nanolaminates on MoS₂ is not suitable for use as a gate dielectric layer. After MoS₂ treated with UV-O₃ treatment, the leakage dots for both O₃-based and H₂O-based La₂O₃/Al₂O₃ nanolaminates decreased. In particular, no leakage dot is observed for O₃-based La₂O₃/Al₂O₃ nanolaminates. It indicates that, with the help of UV-O₃ treatment, ultrathin O₃-based La₂O₃/Al₂O₃ nanolaminates on MoS₂ can serve as the gate dielectric due to its good leakage suppression properties.

Figure 2. AFM results of sub-5 nm La₂O₃/Al₂O₃ nanolaminates on MoS₂ with or without UV-O₃ treatment.
The changes in uniformity of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ may be originated from the interface due to the ALD process of growing La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on silicon is well established [17]. To reveal the changes that occurred at the interface, XPS measurements are performed. Figure 4 shows the C$_{1s}$ spectra of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$. There are mainly two peaks in the C$_{1s}$ spectra for all La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$, which are located at binding energies of 283.0 eV and 284.8 eV. These peaks correspond to the metal carbide and adsorbed carbon, respectively [22]. Moreover, the peak intensity of metal carbide in H$_2$O-based ALD process decreases from 30.0 a.t.% to 15.8 a.t.% after MoS$_2$ suffered from UV-O$_3$ treatment, while that of O$_3$-based ALD process decreases from 11.9 a.t.% to 9.8 a.t.. The lowest metal carbide content in O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ with UV-O$_3$ treatment suggests that the initial interfacial reactions are greatly improved. The appearance of metal carbide is an indication that poor interface reactions occur during the ALD process, which can originate from the generation of intermediates or by-products of metal precursors. MoS$_2$ suffered from low-power UV-O$_3$ treatment form the weak chemical bond of S-O on the surface without hampering its electrical performance [15], which can supply the reaction interface groups at the MoS$_2$ surface during the ALD deposition. As a result, the residuals of the metal carbide or its intermediate precursor during the first ALD reaction cycles can be reduced and the roughness of the nanolaminates can be improved.
In order to further confirm the residue in La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$, Figure 5 shows the Al$_{2p}$ spectra of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$. As shown in Figure 5, the Al$_{2p}$ spectra can be fitted to two peaks, which located at the binding energy of ~74.6 eV and 73.8 eV, respectively. 74.6 eV belongs to the Al-O bond, and the lower 73.8 eV is related to carbide [22]. The content of carbide in H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates decreases from 9.66 a.t.% to 3.88 a.t.% after MoS$_2$ treated with UV-O$_3$ treatment, while that of O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates decreases from 2.98 a.t.% to 0.92 a.t.% after MoS$_2$ treated with UV-O$_3$ treatment. The variation of carbide content in La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ is consistent with the C$_{1s}$ results. Due to lack of dangling bonds or nucleation sites on MoS$_2$, the initial reaction of ALD is dependent on weakly physical adsorbed TMA precursors on MoS$_2$ surface. UV-O$_3$ treatment forms the weak S-O bonds on MoS$_2$ and facilitates the uniform physical adsorption of precursor, which is beneficial for the improvement of initial ALD self-limiting surface reactions. O$_3$ has a stronger ability than water to split the C-H or Al-C bonds which attached to metal atoms in the deposition [18]. As a result, the concentration of metal carbide in O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates is lower than H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates.

Figure 4. C$_{1s}$ spectra of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$.
To determine the valence band offset (VBO) between La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and MoS$_2$, the Kraut method is used which discussed in ref. [23],

$$\Delta E_{VBO} = \frac{E_{Mo^{3p}_{CL}} - E_V^{\text{bulk,MoS}_2}}{E_{Al^{2p}_{CL}} - E_V^{\text{thick nanolaminates}}} - \frac{E_{Mo^{3p}_{CL}} - E_{Al^{2p}_{CL}}}{E_{Mo^{3p}_{CL}} - E_{Al^{2p}_{CL}}}$$

(1)

where $E_{Mo^{3p}_{CL}}$ and $E_{Al^{2p}_{CL}}$ is the binding energy of the Mo$_{3p}$ and Al$_{2p}$ shallow core levels, respectively. $E_V$ is the binding energy corresponding to the valence band maximum (VBM). The value of VBM is determined by the intercept of the slope at the leading edge of the valence band spectrum with the base line. To correct the differential charging, the binding energy calibration was performed using a gold standard sample. Figure 6 shows the core level spectra of ~10 nm sputtered MoS$_2$ with or without UV-O$_3$ treatment. The energy difference between the Mo$_{3p}$ core level and the VBM is 394.58 eV and 394.59 eV for the clean MoS$_2$ and MoS$_2$ treated with UV-O$_3$ treatment, respectively. These values are agreed well with the values reported in ref. [24].

Figure 7 shows the XPS core level spectra of Mo$_{3p}$ and Al$_{2p}$ for La$_2$O$_3$/Al$_2$O$_3$ nanolaminates. The core level energies are obtained by curve fitting to ensure high accuracy binding energy of the peak. In order to measure the band offset between La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and MoS$_2$, ~10 nm H$_2$O-based and O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates are prepared for use as bulk films, respectively. As shown in Figure 7, the energy difference values between the core level energies are determined. Using these energy difference values with Equation (1), the VBO values of the H$_2$O-based and O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ can be derived. The VBO of 3.10 eV and 3.14 eV is obtained for O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ and MoS$_2$ with UV-O$_3$ treatment, respectively.
tively. In addition, the VBO of H$_2$O-based nanolaminates on MoS$_2$ and MoS$_2$ with UV-O$_3$ treatment is 2.75 eV and 2.91 eV, respectively. The results indicate that the VBO is affected by the different oxidants and UV-O$_3$ treatment. The negligible VBO variations for O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates suggest that it has a better stability compared to H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates.

![Figure 6](image-url) 

Figure 6. The XPS core level spectra of Mo$^{3p}$ for MoS$_2$. 

To obtain the conduction band offset (CBO) between La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and MoS$_2$, the optical band gaps of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates are measured. The optical band gaps form the plots of $(aE)^2$ versus photo energy $E$ are shown in Figure 8. The extrapolation of the linear part of $(aE)^2 - E$ down to $(aE)^2 = 0$ gives the values of band gaps [25]. The measured band gap value of O$_3$-based and H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates is 6.37 eV and 6.19 eV, respectively. These values are in good agreement with the reported values of La$_2$O$_3$/Al$_2$O$_3$ gate stack or LaAlO$_3$ films ranging from 6.1–6.4 eV [17,26]. The results indicate that the O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates has a larger bandgap value compared to the H$_2$O-based nanolaminates. This may be caused by the lower content of impurities found in O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates compared to H$_2$O-based nanolaminates.
The XPS core-level and valence band spectra of thin and bulk La$_2$O$_3$/Al$_2$O$_3$ nanolaminates. 

Figure 7. The XPS core-level and valence band spectra of thin and bulk La$_2$O$_3$/Al$_2$O$_3$ nanolaminates.

Figure 8. $(\alpha E)^2$ versus photo energy $E$ of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates.
Using the calculated VBO and band gap values, the conduction band offset between La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and MoS$_2$ can be attained by the following equation:

$$\Delta E_{\text{CBO}} = E_{g}^{\text{La}_2\text{O}_3/\text{Al}_2\text{O}_3 \text{ nanolaminates}} - E_{g}^{\text{MoS}_2} - \Delta E_{\text{VBO}}$$  (2)

where $E_{g}^{\text{La}_2\text{O}_3/\text{Al}_2\text{O}_3 \text{ nanolaminates}}$ and $E_{g}^{\text{MoS}_2}$ is the bandgap of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and MoS$_2$, respectively. The bandgap of 1.4 eV for MoS$_2$ is used here [27]. According to the Equation (2), the CBO of O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ and MoS$_2$ with UV-O$_3$ treatment is 1.87 eV and 1.83 eV, respectively. Meanwhile, the CBO of H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ and MoS$_2$ with UV-O$_3$ treatment is 2.04 eV and 1.88 eV, respectively. The corresponding band diagrams are illustrated in Figure 9. It can be seen that both La$_2$O$_3$/Al$_2$O$_3$ nanolaminates/MoS$_2$ interface have a Type I alignment, where the conduction band edge and valence band edge of MoS$_2$ are located within the bandgap of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates. Furthermore, both CBO and VBO values of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ provide excellent electron and hole barriers due to their values larger than 1 eV, ensuring La$_2$O$_3$/Al$_2$O$_3$ nanolaminates suitability for FETs applications. Remarkably, O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates has a higher VBO compare with H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates, which is better for p-channel FETs application.

![Figure 9](image_url)

**Figure 9.** Band diagrams of (a) O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates and (b) H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ (Blue for MoS$_2$ and red for MoS$_2$ treated with UV-O$_3$ treatment).

Figure 10 shows the I-V curves of La$_2$O$_3$/Al$_2$O$_3$ nanolaminates on MoS$_2$ after fabricated metal-oxide-semiconductor (MOS) capacitor. At the applied voltage of 2 V, for O$_3$-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates, the leakage current decreased from $1.2 \times 10^{-2}$ mA to $9.6 \times 10^{-3}$ mA, while the breakdown voltage increased from 9.01 V to 10.21 V after MoS$_2$ treated with UV-O$_3$ treatment. The same trend is observed in H$_2$O-based La$_2$O$_3$/Al$_2$O$_3$ nanolaminates. The leakage current decreased from $2.6 \times 10^{-2}$ mA to $2.3 \times 10^{-2}$ mA, while the breakdown voltage increased from 6.76 V to 7.36 V after MoS$_2$ treated with UV-O$_3$ treatment. The breakdown voltage is obtained when the leakage current reaches 1 mA [28]. The deceased of leakage current and increase of breakdown voltage may be attributed to the uniformity of the La$_2$O$_3$/Al$_2$O$_3$ nanolaminates as well as the reduction of impurities or residuals at the interface. The leakage current may originate either Poole-Frenckel or Fowler-Nordheim mechanism from the point of view of quantum tunneling [29,30], which has been confirmed in our measurement. The lowest leakage current and highest break-
down voltage are obtained for O3-based La2O3/Al2O3 nanolaminates on MoS2 with UV-O3 treatment, making it a promising dielectric candidate for the application of MoS2 FETs.

4. Conclusions

In this study, atomic layer deposition growth of sub-5 nm La2O3/Al2O3 nanolaminates on MoS2 using different oxidants (H2O and O3) and the UV-O3 pretreatment on MoS2 are investigated. Compared with H2O-based La2O3/Al2O3 nanolaminates on MoS2, better uniformity and lower leakage dots were observed for O3-based La2O3/Al2O3 nanolaminates on MoS2. This is associated with the metal carbide concentration in La2O3/Al2O3 nanolaminates on MoS2, which is generated by insufficient interfacial reactions. UV-O3 treatment can decrease the residuals of the metal carbide and improve the deposition of La2O3/Al2O3 nanolaminates on the MoS2 interface by introducing the weak S-O bonds to MoS2 surface, leading to the properties of La2O3/Al2O3 nanolaminates being substantially improved. The band offset values of both O3-based and H2O-based La2O3/Al2O3 nanolaminates/MoS2 are larger than 1 eV, which can provide eligible electron and hole barrier height. In particular, a higher valence band offset is obtained for O3-based La2O3/Al2O3 nanolaminates compared to H2O-based La2O3/Al2O3 nanolaminates. Consequently, O3-based La2O3/Al2O3 nanolaminates on MoS2 exhibits smaller leakage current and higher breakdown voltage, especially after MoS2 suffered from UV-O3 treatment. All results indicate that O3-based La2O3/Al2O3 nanolaminates on MoS2 with UV-O3 treatment is a more appropriate process to obtain sub-5 nm uniform La2O3/Al2O3 nanolaminates on MoS2 due to its good electrical characteristics, providing important implications for its integration into transistors.

Author Contributions: J.F. performed the data analyses and wrote the manuscript; Y.S., performed the experiment; H.L., S.W. and L.D. helped perform the analysis with constructive discussions; L.L., Y.Z. and X.W. contributed to the conception of the study. All authors have read and agreed to the published version of the manuscript.

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