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Impact of Radiation Effect on Ferroelectric Al-Doped HfO_2 Metal-Ferroelectric-Insulator-Semiconductor Structure

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ABSTRACT The γ-ray total dose radiation effects on ferroelectric Al-doped HfO_2 (HfAlO) thin films with an n-type Si substrate were studied. The I-V, P-V, C-V and fatigue characteristics of the HfAlO-based Metal-Ferroelectric-Insulator-Semiconductor (MFIS) structure were analyzed with the increasing total dose from 0 to 1 Mrad (Si). The remnant polarization (P_r) values, the capacitance (C) values and the switching voltage (V_c) of this MFIS gate structure decreased with the increasing total dose. A TCAD model of Metal/HfAlO/SiO_2/Si (MFIS) was proposed to explain the degradation mechanism of ferroelectric properties of the MFIS structure. We find that the new interface defects caused by radiation can reduce the electric field strength with the increasing total dose, which can significantly influence the irradiation properties of the HfAlO/SiO_2/n-Si gate structure. These results provide the basis to applications for the HfO_2-based devices in radiation working environment.

INDEX TERMS Metal-ferroelectric-insulator-semiconductor (MFIS), Hafnium Aluminium Oxide (HfAlO), remnant polarization, γ-ray radiation, ferroelectric memory.

I. INTRODUCTION

Nowadays the most commercial of nonvolatile memory (NVM) available are semiconductor Flash-based memory. However, the equivalent gate oxide thickness (EOT) of the gate stack in Flash memory is limited to 8 nm, which significantly influences its physical scalability and storage capacity [1]. The conventional flash memory would be replaced by ferroelectric field-effect transistor (FeFET) memory, which was one of the most promising non-volatile memory [2]. Since perovskite-based ferroelectrics material such as (Pb, Zr)TiO_3 (PZT) for FeFET memory have a relatively high permittivity with values about few hundreds, the stack structure among the depletion layer in the silicon, the ferroelectric layer and the interface oxide brings about a high depolarization field, leading to a poor retention characteristics [3]. The doped HfO_2 thin films with a thickness below 10 nm have the advantages of high scalability, excellent retention, mature production and good CMOS compatibility, which are considered to be the best replacement for the traditional ferroelectric materials of FeFET in information storage fields [4]–[6]. Besides, HfO_2-based ferroelectric capacitor memories show higher radiation immunity as compared with that of traditional non-volatile memories [7].

HfO_2-based ferroelectric capacitors were proved to be a high radiation tolerance [8]–[10]. Y-doped HfO_2-based

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ferroelectric capacitors have a radiation-hardened stability above the total dose of 10 Mrad (Si) [8]. Zr-doped HfO_2-based ferroelectric capacitors also display a robust ferroelectric performance against radiation with a total dose of 1 Mrad (Si) [9]. The remnant polarization (P_r) of Al-doped HfO_2 (HfAlO) ferroelectric capacitors have been reported to have a little decrease by 9.2% from 0 to 5 Mrad (Si) in our recent work [10]. However, the radiation mechanisms of HfO_2-based FeFET or Metal-Ferroelectric-Insulator-Semiconductor (MFIS) structure have not been clearly understood yet. Our previous work showed that irradiation-induced electrical characteristics degradation of MFIS structure of $Pt/SrBi_2Ta_2O_9/HfTaO/Si$ were found under the total dose of 10 Mrad (Si) [11]. D. J. McCrory *et al.* verified the defects induced by γ-ray radiation to be an O_2^- coupled to a hafnium ion of $TiN/Ti/HfO_2/TiN$ RRAM devices through electrically detected magnetic resonance (EDMR) [12]. It has verified that interface defects have been formed in the $TiN/HfO_2/Si$ MOS capacitor during irradiation, and the accumulation capacitance decreased with the increasing of irradiation dose [13]. Brewer *et al.* indicated that the radiation effect can be also influenced by orientation, domain size, grain boundary density and morphology [14], [15]. Palade *et al.* [16] demonstrated that the trilayer of control HfO_2/floating gate of Ge nanoparticles in HfO_2/tunnel HfO_2/Si substrate containing HfO_2 nanocrystals with the orthorhombic/tetragonal structure has a high sensitivity of 0.8 mV Gy^{-1} to α particle irradiation. Chen *et al.* [17] studied the impact of γ-ray radiation on the performance of Zr-doped HfO_2-based FeFET memory on Si, and illustrated that hole trapping from radiation-induced defects caused the adverse effect for endurance performance. However, the radiation mechanisms on the HfAlO-based MFIS structure with an n-type Si substrate is still missing.

Herein, the γ-ray radiation effects on HfAlO MFIS structure and the relevant electrical performance degradation mechanisms after radiation were investigated. A Technology Computer Aided Design (TCAD) simulation was used to explain how the interface defects influence the performances of the MFIS structure.

II. EXPERIMENTAL

HfAlO thin films were deposited on an N-doped Si(100) substrate via thermal ALD at 300 °C. The resistivity of the Si substrate is 0.005 Ωcm. Here, ALD cycle ratio of $HfO_2:AlO_{3/2}$ was set to 39:1. The growth rates of HfO_2 and $AlO_{3/2}$ at 300 °C are ~1.05 Å/cycle and ~1.1 Å/cycle, respectively. Thus the thickness of HfAlO thin films was estimated to 10 nm with 100 ALD cycles (39 cycles of HfO_2 and 1 cycle of $AlO_{3/2}$ for twice, 20 cycles of HfO_2 for once). TiN was sputtered as the upper electrode with a thickness of 40 nm. The HfAlO samples were annealed at 400 °C for 20 s in nitrogen ambient by rapid thermal annealing (RTA) after the deposition of TiN electrodes. There has an improvement in ferroelectric properties from 350 to 600 °C. For the purpose of CMOS compatibility, we choose the

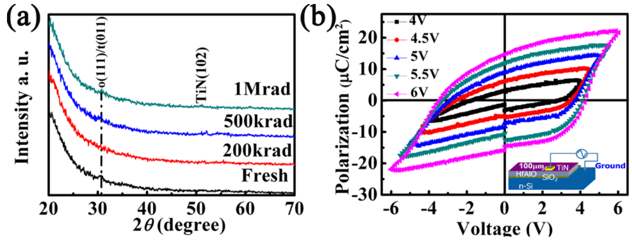

FIGURE 1. XRD patterns and P-V curves: (a) GIXRD spectrum for HfAlO MFIS structure before and after γ-ray irradiation; (b) Ferroelectric polarization electric hysteresis loops under various voltages, the inset was the schedule of electrical performance tests.

samples at low temperature of 400 °C for our manuscript. Pt/TiN top electrodes were produced by etching a square of 100 μm×100 μm via inductively coupled plasma (ICP) method. ^{60}Co γ-ray was used as the radiation source and the radiation rate of this work is set to 80 rad (Si)/s. All of our samples were adhere to the PCB board. During the radiation, an open circuit state for devices were adopted and the incident direction of γ-ray was 90 degrees from the sample surface. Dry ices were used to store our samples after γ-ray irradiation, and all of performance measurements were measured at the same temperature within 72 hours to avoid the annealing effect of radiation. The total doses of 200 krad, 500 krad and 1 Mrad were used to our three similar groups. Grazing-angle Incidence X-ray Diffraction (GIXRD, incidence angle of 1°) was used to analyze the crystal structures of the HfAlO films on Si substrate. Scanning Transmission Electron Microscopy (STEM) and High-resolution Transmission Electron Microscopy (HRTEM) were adopted to investigate the cross-sectional specimens with the help of a dual beam focus ion beam system (FIB, Quanta 3D, FEG). Semiconductor device analyzer (Agilent, USA, B1500A) was used to study the current density and capacitance of the MFIS samples. The polarization-voltage (P-V) and fatigue characteristics were gained through the Ferroelectric test systems (USA, Radiant Technologies Precisions Workstations). The model of ferroelectric electric hysteresis loops is available within the Sentaurus Device Simulator of Synopsis TCAD.

III. RESUITS AND DISSCUSSIONS

GIXRD spectrum for HfAlO MFIS structure under the radiation dose from fresh (0) to 1 Mrad(Si) was displayed in Fig. 1(a). All the samples were measured after the annealing process. The weak peak centered at 30.6° in HfAlO MFIS structures was assigned to the tetragonal phase (t-phase) or the non-centrosymmetric orthorhombic phase (o-phase), which are unstable and responsible for the ferroelectric properties of HfO_2-based thin films [18], [19]. Except the peaks of the o-phase, there is no any other diffraction peak shown up after the HfAlO thin film deposited, suggesting that this MFIS structure is mostly amorphous, which corresponds to the results of the HRTEM in Fig. 2(c). Fig. 1(b) presents the P-V hysteresis curves for HfAlO MFIS structure. The inset was the schedule of electrical performance tests. The bottom

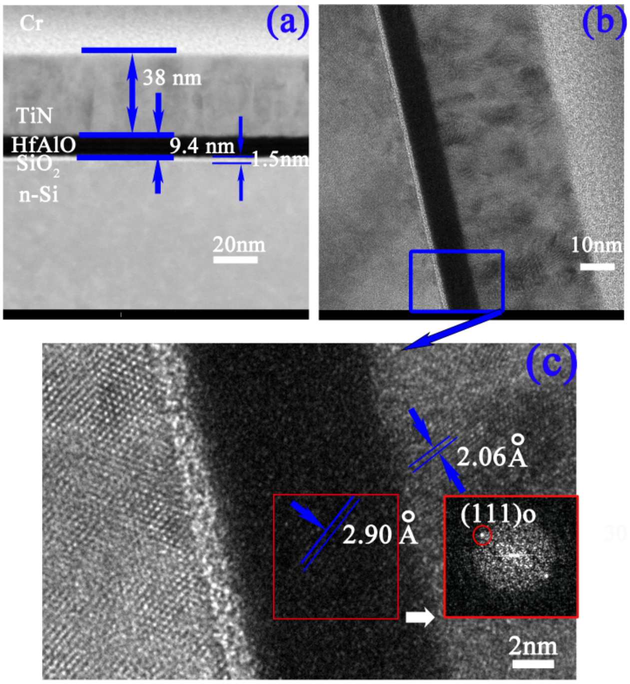

FIGURE 2. Microstructure characterizations: (a) Z-contrast STEM image of the HfAlO MFIS sample; (b) and (c) Cross-sectional HRTEM images and the enlarged image of the selected area of MFIS structure, respectively.

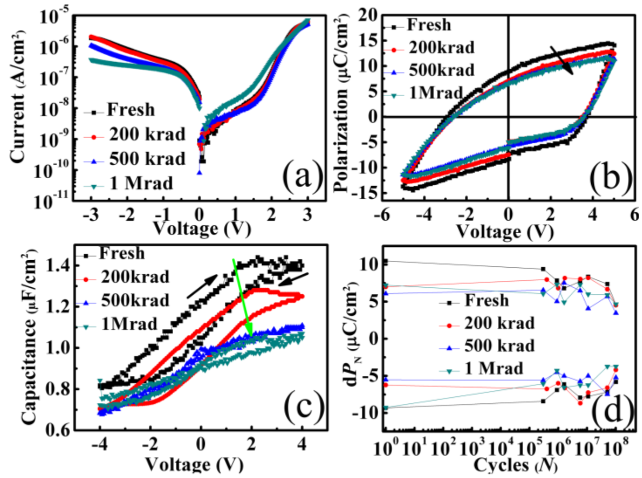

FIGURE 3. ***I-V***, ***P-V***, ***C-V*** **and fatigue characteristics of the HfAlO MFIS structure before and after γ-ray irradiation: (a)** ***I-V*** **curves; (b)** ***P-V*** **curves, (c)** ***C-V*** **curves measured at 1 MHz; (d) Fatigue characteristics.**

electrode of Si substrate was grounded in the electrical measurements. It can be seen that the remnant polarization (P_r) values of the MFIS structure get larger with the increasing voltages. $2P_r$ and $2V_c$ can be expressed as "$2P_r = P_r^+ - P_r^-$" and "$2V_c = V_c^+ - V_c^-$", respectively, where P_r^+ or P_r^- represents remnant polarization under forward or negative voltage sweep, V_c^+ or V_c^- shows the switching voltage under forward or negative voltage sweep. The values of $2P_r$ and $2V_c$ of the MFIS structure under the voltage of 6 V were 30.9 μC/cm^2 and 7.7 V, respectively. The ferroelectric properties of this similar structure with an n-type Si substrate have also been reported by Liu *et al.* [20]. It can be implied that the amounts of ferroelectric domains have been increased with the larger voltages.

Fig. 2(a) presents the *Z*-contrast STEM image of the HfAlO MFIS structure. Thickness of SiO_2, HfAlO, and TiN were 1.5 nm, 9.4 nm and 38 nm, respectively, while the thickness of HfAlO thin film and TiN layer corresponds to the estimated thickness of 10 nm and 40 nm as mentioned in the experiment part. Figs. 2(b) and 2(c) display the cross-sectional HRTEM images and the enlarged image of the selected area of MFIS structure, respectively. The inset of the areas marked by a red rectangle in Fig. 2(c) was the fast fourier transformation (FFT) diffractogram, which certifies the orthorhombic phase (111) plane with an interplanar distance of 2.90 Å, implying the formation of ferroelectric phase [21], [22]. Moreover, (111) plane of TiN with an interplanar distance of 2.06 Å which corresponds to (111)-oriented plane of TiN was discovered at the interface between HfO_2 (ferroelectric phase) and TiN. It can be speculated that the formation of the ferroelectric phase of HfAlO thin films is related to the interface of (111)-oriented plane of TiN.

Fig. 3 depicts the *I-V*, *P-V*, *C-V* and fatigue characterizations of the HfAlO MFIS structure before and after γ-ray irradiation. We can clearly see that the leakage current shows a large decrease under negative voltage with the irradiation from 0 to 1 Mrad (Si), as shown in Fig. 3(a). This phenomenon may be due to the radiation-induced trapped holes in the oxide that forms smaller hole concentration at the interfaces of HfAlO/SiO_2. However, the current displays an increase trend in the range of 0 to 1 V, and then shows irregular change in the range of 1 to 3 V as the radiation increases from 0 to 1 Mrad (Si). The increase under positive bias may be due to the radiation-induced trapped holes in the oxide that causes higher electron concentrations at the top TiN electrode. More interface defects can be induced with an increase to the dose of 1 Mrad [13]. The values of P_r and the capacitance in *C-V* curves of the HfAlO MFIS structure decrease with the increasing dose from fresh to 1 Mrad as represented in Figs. 3(b) and 3(c), which implicated that the defects induced by the larger total dose of radiation have affected enhanced pinned domains and dielectric loss. The fatigue characteristics under radiation environment were displayed in Fig. 3(d). The pulse amplitudes were 1.8 V for fatigue tests of the HfAlO MFIS structure. The dP_N can be described as the difference between the non-switched remnant polarization and the switched remnant polarization after *N* cycles pulse switching. The reductions of dP_N were 46.75%, 33.50%, 24.25% and 48.25% with an increase dose from fresh to 1 Mrad. The fatigue characteristics become better after radiation from 0 to 500 krad, and get worse from 500 krad to 1 Mrad. The interface defects trapping effect may be responsible for this phenomenon. With the increasing dose from 0 to 1 Mrad, the new interface defects at the interface of HfAlO/SiO_2 can reduce the potential difference

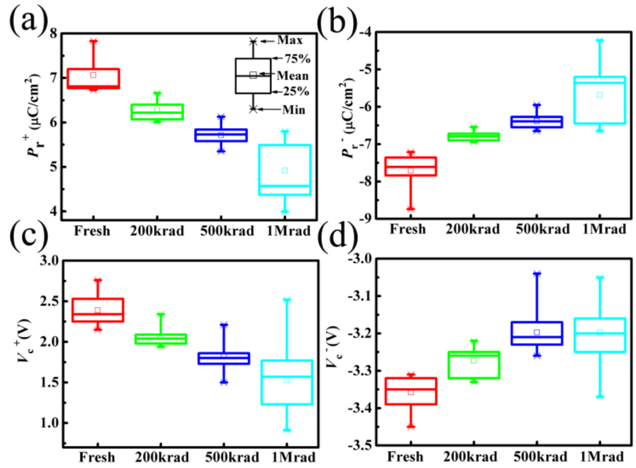

FIGURE 4. Statistical P_r^+, P_r^-, V_c^+ and V_c^- values of the HfAlO MFIS structure before and after radiation for (a), (b), (c) and (d), respectively.

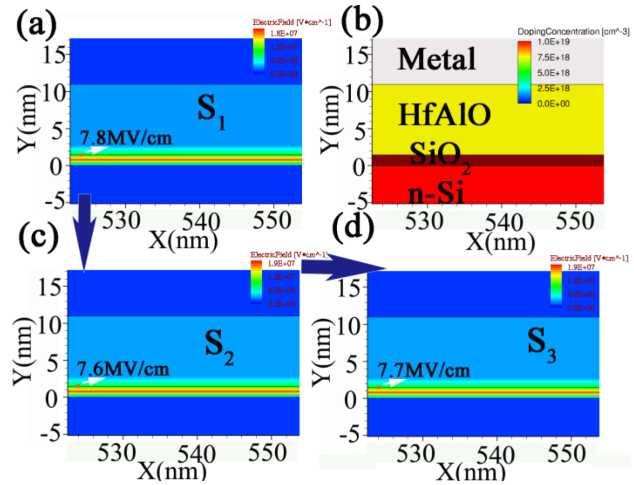

FIGURE 5. The TCAD structure schematics: (b) the TCAD structure of Metal/HfAlO/SiO_2/Si (MFIS); (a), (c) and (d) represents the state of S_1, S_2 and S_3, respectively.

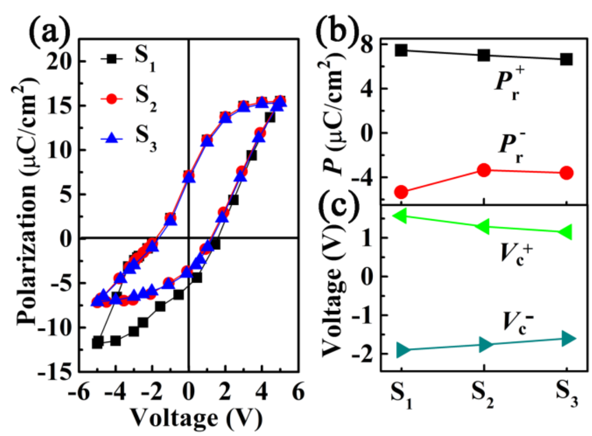

FIGURE 6. TCAD simulation results: (a) the P-V curves of S_1, S_2 and S_3; (b) P_r^+ and P_r^- changes from S_1 to S_3; (c) V_c^+ and V_c^- changes from S_1 to S_3.

between the upper and bottom electrodes. Then the weakened defects trapping effect can be the main factor to slow down the decay rate of P_r^+ and V_c^- with the dose from 500 krad to 1 Mrad.

To further study the radiation effects on ferroelectric properties of the HfAlO MFIS structure, the P_r^+, P_r^-, V_c^+ and V_c^- of 4 groups (20 devices for each group) were tested before and after γ-ray radiation. These statistical box charts of P_r^+, P_r^-, V_c^+ and V_c^- values of the HfAlO MFIS structure under total dose from fresh to 1 Mrad were counted in Fig. 4. The values distribution was expressed by a shape of a box chart, and the detail percentile was marked in the inset of Fig. 4(a). As the total dose increases from fresh to 1 Mrad, the mean of P_r^+ and P_r^- of the HfAlO MFIS structure decrease by 56.52% and 26.0% from fresh to 1 Mrad (6.9 to 3.0 μC/cm^2, -7.7 to -5.7 μC/cm^2), respectively, as presented in Figs. 4(a) and 4(b). As the total dose of the γ-ray radiation increases, more movable charges or charged domain walls can be compensated by radiation oxide trapped defects or interface defects, while these movable charges or charged domain walls can also take part in the polarization switching for ferroelectric thin films as reported by Placeres-Jiménez *et al.* [23]. Thus, the increasing trapped centers can reduce the probability of polarization switching in the HfAlO MFIS structure. There are also similar change trends for switching voltages. Absolute values of V_c^+ and V_c^- of the HfAlO MFIS structure decrease by 36.55% and 4.76% with an increase of dose from fresh to 1 Mrad as shown in Figs. 3(c) and 3(d). The difference shown in the change of V_c^+ and V_c^- values with the increasing dose may be due to the increasing concentrations of the positive radiation-trapped holes or oxygen vacancies in the oxide of HfAlO. Therefore, the V_c^+ can be reduced more significantly caused by the decreased potential barrier under forward voltage sweep as discussed in our previous work [24].

To explain the radiation effects on the MFIS structure, a Metal/HfAlO/SiO_2/Si (MFIS) model was proposed by Sentaurus Device Simulator of Synopsis TCAD [25]. Coercive fields used within the ferroelectric model were set to 1.5 MV/cm, while the saturated polarization and the remnant polarization of HfAlO thin films were set to 20 and 12 μC/cm^2, respectively. The thickness of HfAlO and SiO_2 layer were set to 9.4 nm and 1.5 nm, respectively. The TCAD structure schematic, evolution of electric field and simulation results were shown in Fig. 5 and Fig. 6. According to our previous work [26] and the reference 27, the trap density (N_{it}) at the HfAlO/SiO_2 interface and the trap density in HfAlO were first set to 10^{10}cm^{-3} and 10^{18}cm^{-3}, which was called the fresh state of S_1. The defects induced by radiation can first increase a part of trapped charges such as oxygen vacancies in the HfAlO layer, and enhance the domain pinned effect, which was called S_2 with charge concentration of 10^{10} cm^{-3} and 10^{19}cm^{-3} for HfAlO/SiO_2 interface and HfAlO layer, respectively. With the increasing total dose, more defects can be induced at HfAlO/SiO_2 interface, which is called S_3 with a concentration of 10^{11} cm^{-3} and 10^{19} cm^{-3} for HfAlO/SiO_2 interface and HfAlO layer, respectively. It can be seen that the P_r^+, P_r^-, V_c^+ and V_c^- values decreased with the state changing from S_1 to S_3, which corresponds well with our experimental results induced by the increasing total dose as shown in Fig. 6. The evolution process for the radiation effects on Metal/HfAlO/SiO_2/Si (MFIS) may follow the state conversation of "$S_1 \rightarrow S_2 \rightarrow S_3$". First, a part of oxygen vacancies was induced by radiation in the HfAlO layer as shown in Fig. 5(b), which can inhibit the nucleation of ferroelectric domains and polarization switching, resulting a reduction for the polarization. Then the electric field strength at the interface may reduce with an amount of defects induced by radiation as shown in Figs. 5(a) and 5(c). With the increasing dose from 500 krad to 1 Mrad, the increasing interface defects of HfAlO/SiO_2 can further influence the ferroelectric properties and slow down the decay rate of P_r^+ and V_c^-. Thus, the defects originated from both the HfAlO layer and the interface of HfAlO/SiO_2 have determined the electrical performance under the radiation.

IV. CONCLUSIONS

The γ-ray total dose radiation effects on HfAlO MFIS structure has been investigated. The values of P_r and the highest capacitance in C-V curves of the HfAlO MFIS structure decrease with the increasing dose from fresh to 1 Mrad due to the enhanced pinned domains and dielectric loss. A Metal/HfAlO/SiO_2/Si (MFIS) TCAD model was proposed to simulate the radiation effects. The defects induced by radiation such as oxygen vacancies in the HfAlO layer can influence the ferroelectric properties under the total dose from 0 to 500 krad. With the increasing dose from 500 krad to 1 Mrad, the increasing interface defects at the HfAlO/SiO_2 interface can slow down the decay rate of P_r^+ and V_c^-. The research results can be helpful to understand radiation mechanisms of MFIS structure and promote the applications of HfO_2-based FeFET devices under radiation environment.

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