# Supporting Information

**Title:** Impact of radiation effect on energy storage density and wake-up behaviors of antiferroelectric-like Al-doped $HfO_2$ thin films

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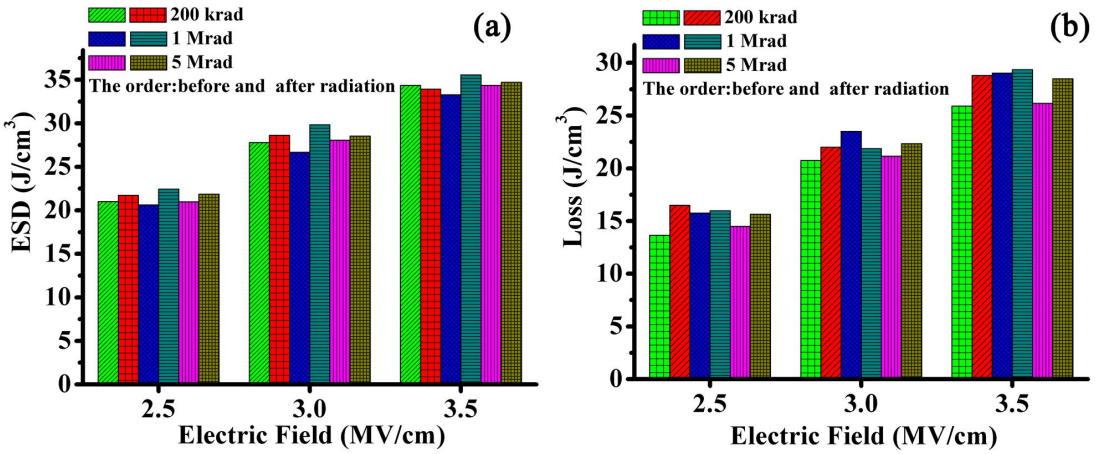


**Figure S1.** a) the sum of energy storage density (EDS) and loss of the AFE-like HfAlO thin films before and after radiation ; b) the loss of the thin films before and after radiation from 200 krad to 5 Mrad.

As discussed in the main paper, Figure S1 shows the ESD and loss of the AFE-like HfAlO thin films before and after radiation. Under the three total doses, the EDS of all the samples shows an increase after the radiation. What's more, the loss of the thin films also shows a relatively increment under the dose of 200 krad and 5 Mrad. The circumstance of 1 Mrad has an irregular change, which might be due to the annealing effect for the first day to do the radiation tests for the total dose of 1 Mrad.

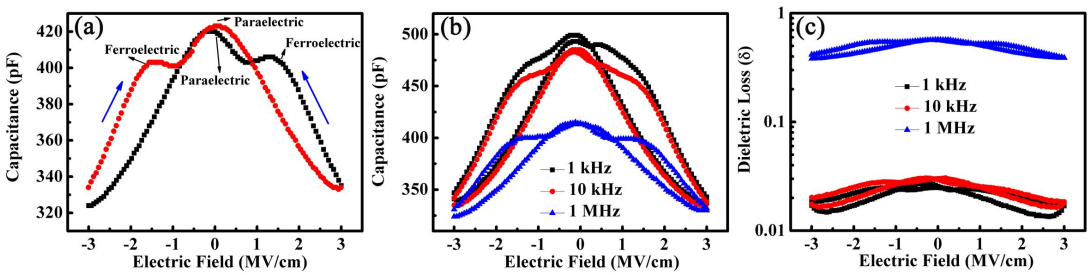


**Figure S2.** a) the *C-V* switching characteristics of AFE-like HfAlO thin films; b) the *C-V* curves of the AFE-like HfAlO thin films under the measuring frequency from 1 kHz to 1 MHz; c) the frequency dependent dielectric loss curves of the thin films.

To elucidate the switching directions of *C-V* curves, Fig. S2(a) represents the *C-V* curves from -3 to +3V (red circles) and from +3 V to -3 V (black rectangle), respectively. The films show a phase transition from ferroelectric phase to paraelectric phase as the maximum appears around the zero in the *C-V* curves. Fig. S2(b) exhibits the *C-V* curves under different measuring frequency. As can be seen, the values of the capacitance decreased with the increasing measuring frequencies. In the low-frequency *C-V* measurements, the domain switching can contribute a part for the *C-V* response. With the increasing frequency, the contributions of the domain switching should get weaker, while the other polar elements (space charges, ionic polarization, reversible domain wall motion, and lattice fluctuations) play a greater role [1]. Therefore, the values of capacitance at high frequency would get smaller by the weak response of domain switching. Meanwhile, the dielectric loss curves also showed that the space charge effect had a greater influence with the increasing measuring frequency.

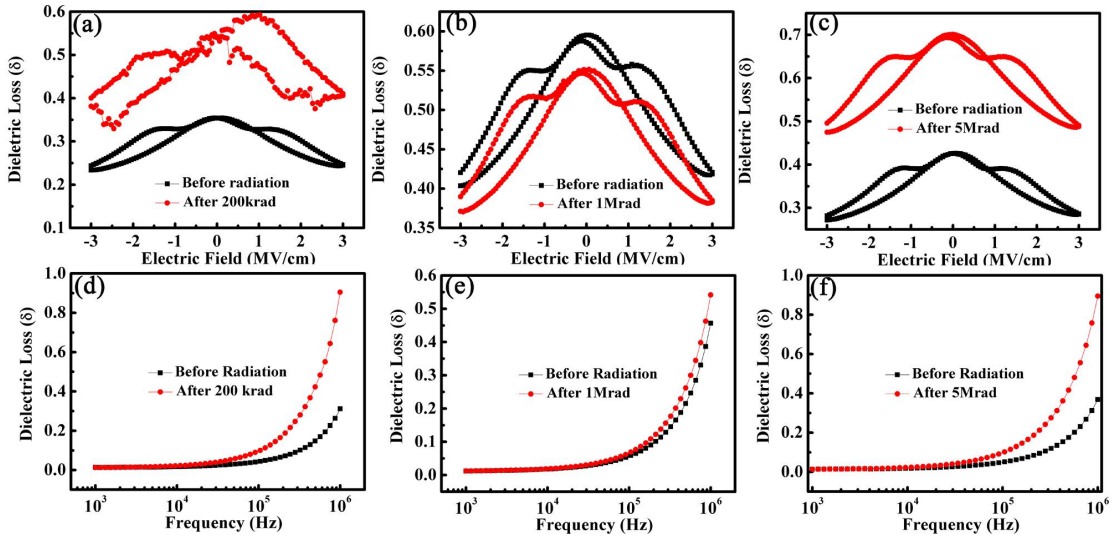


**Figure S3.** Dielectric loss (*D*)-*V*, and D-*f* curves of AFE-like HfAlO thin films before and after γ -ray irradiation at the same electrodes: (a) and (d) before and after 200 krad (Si); (b) and (e) before and after 1 Mrad (Si); (c) and (f) before and after 5 Mrad (Si), respectively.

As mentioned in the main paper, the dielectric loss(*D*)-*V* curves and *D*-*f* curves had also been depicted before and after the radiation as shown in Figure S3. With the increase of the total dose from 200 krad to 5 Mrad, an increasing dielectric loss can be found, which was similar to the frequency dependent dielectric loss curves as shown in Fig. S2(c). It was further verified that more trapped defects and less switchable domains should be produced by the radiation. As can be seen, a relative increase can be found especially at the frequency higher than 100 kHz, which might be due to the increasing space charges or the interface traps at the interfaces of TiN/HfAlO.

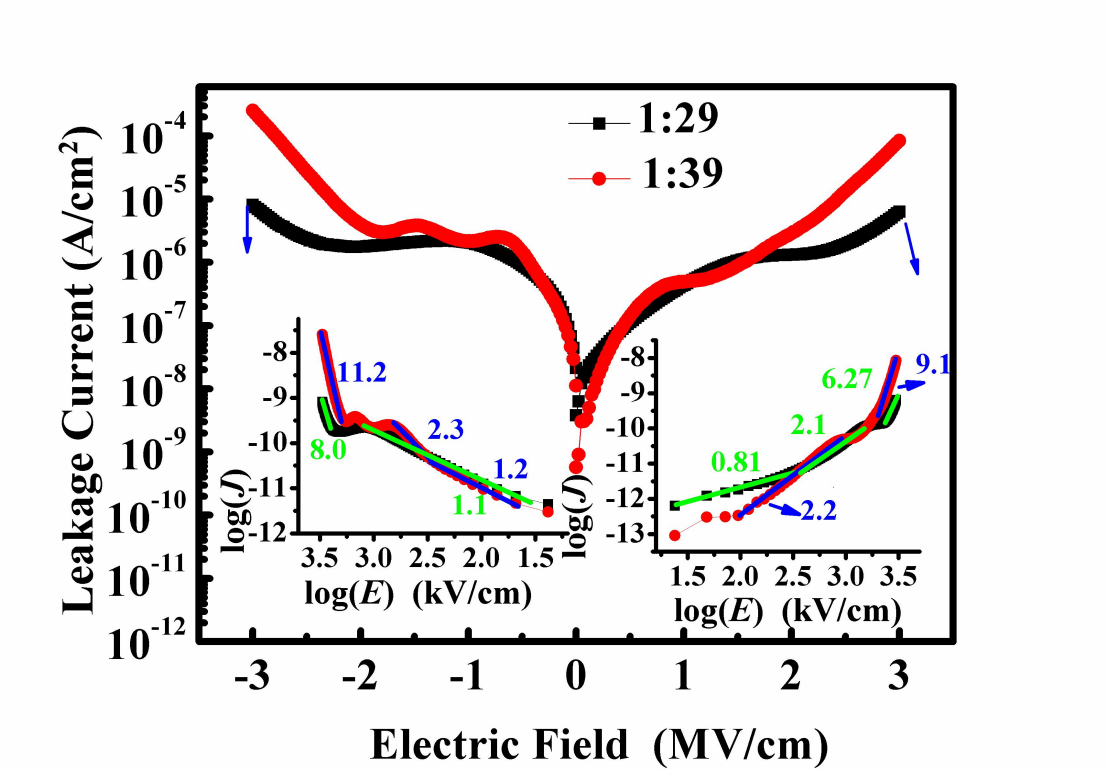


**Figure S4.** Comparisons of *J-E* curves of AFE-like HfAlO (29:1) and FE HfAlO (39:1) thin films. The insets in the Figure S4 show log*J* versus log*E* characteristic and the linear fit for the slop.

Interestingly, as discussed in the main paper, there were quite different from the regions with the slopes much larger than 2 for AFE-like HfAlO (29:1) and FE HfAlO (39:1) thin films. As can be seen in Figure S4, the FE thin films show a relatively higher leakage current than the AFE-like thin films in the the polarization modulated Schottky contact regions [2], which can be due to a higher ferroelectric polarization in FE thin films. The Ohm's law region in FE thin films are quite shorter than that in AFE-like thin films, which also suggests that more interface trapped defects between the TiN and HfAlO should exist in the FE thin films, leading to an earlier SCLC current of FE thin films.

[1] C. Brennan, *Integr. Ferroelectr.* **9**, 335-346 (1995).

[2] F. Yang, F. W. Zhang, G. D. Hu, Z. H. Zong, and M. H. Tang, *Appl. Phys. Lett.* **106**, 172903 (2015).