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***Research article***

**Demonstration of ferroelectricity in PLD grown HfO2-ZrO2 nanolaminates**

**Sree Sourav Das1,\*, Zach Fox1, Md Dalim Mia2, Brian C Samuels2, Rony Saha2, and Ravi Droopad1,2**

**1** Ingram School of Engineering, Texas State University, San Marcos, TX-78666, USA

**2** Materials Science, Engineering and Commercialization, Texas State University, San Marcos, TX-78666, USA

**\* Correspondence:** Email: xky3tv@virginia.edu; Tel: +1-737-213-3051.

**Supplementary**

**1. High resolution XRD scan**

Since our deposited films were polycrystalline, we did not perform asymmetric scan. Hence only XRD 2θ scans were performed to evaluate the crystal structure. To reduce noise, normalized XRD spectra of the samples are plotted below focusing a small window. We compared the obtained peak position of different phases with other experimental results ([21,24] in the manuscript) and found a good match with them. We also confirmed the presence of orthorhombic phase using Raman spectroscopy from vibrational peak shift.

Chart

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**Figure S1.** Normalized HRXRD 2θ scan of HfO2-ZrO2 deposited at 750 °C and Po= 1.2 × 10−4 torr for various layer thicknesses.

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**Figure S2.** Normalized HRXRD 2θ scan of 15 nm HfO2-15 nm ZrO2 deposited at 750 °C for various oxygen pressure.

Chart, line chart

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**Figure S3.** Normalized HRXRD 2θ scan of 15 nm HfO2-15 nm ZrO2 deposited at Po= 1.2 × 10−4 torr for various temperatures after annealing.

**2. HfO2-ZrO2 Interface**

To examine the interface between HfO2 and ZrO2, XPS depth profile measurements were taken in every 2 second interval during ion etching of the sample from the surface to the bottom of TiN electrode. An argon ion source having energy 4000 eV as used for etching. Figure S4 show the profile of HfO2 and ZrO2 signal for 2 structures grown at different temperatures. From region 1, only signal of ZrO2 is obvious; signal from other elements is entirely absent. As etching continues, i.e., in region 2, ZrO2 signal begins to decay while signal of ZrO2 is totally absent and HfO2 appears as the dominant signal. Finally, further etching reveals the bottom electrode TiN. On the other hand, in case of 750 °C deposition temperature, ZrO2 shows a significant amount of atomic percentage even in region 3. That means ZrO2 diffused into the HfO2 layer. However, the composition of HfO2 and ZrO2 in the intermixing layer is difficult to determine because of the penetration depth of X-rays (~10nm). Transmission electron microscopy (TEM) and Secondary Ion Mass Spectroscopy (SIMS) can be better alternative to validate the compositional percentage of the intermixing layer.

Diagram

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**Figure S4.** Etching profile of 15 nm HfO2/15 nm ZrO2. Atomic percentage of different compound vs etching time from surface to bottom electrode.

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