Supporting Information

Effect of Hf content on the microstructure and ferroelectric properties in $Hf_xZr_{1-x}O_2$ thin films by all-inorganic aqueous precursor solution

Jingjing Wang[a], Dayu Zhou[a,*], Wei Dong[a], Ziqi Li[a], Nana Sun[a], Xiaoduo Hou[a], Feng Liu[b]

a Key Laboratory of Materials Modification by Laser, Ion, and Electron Beams (Ministry of Education), School of Materials Science and Engineering, Dalian University of Technology, Dalian 116024, China

*To whom corresponding should be addressed:

* Email: zhoudayu@dlut.edu.cn

b Instrumental Analysis & Research Center, Dalian University of Technology • Panjin Campus, Panjin 124221, China

S1. The hard mask for the top electrode and measurement structure of $Hf_xZr_{1-x}O_2$ (HZO) film

The hard mask design drawing of TiN top electrode was shown in Fig. S1(a).With the aid of a stainless steel mask, 80 nm thick TiN of circular electrode points with the radius of 55 μm, 100 μm and 225 μm, and two big rectangular electrode structures (5500×4000 μm^2) prepared by magnetron sputtering process were fabricated on the surface of the crystalline HZO film. In this work, n-type (100) Si substrate with a lower resistivity (10^{-3}~10^{-2} Ω·cm) was used as the bottom electrode. During the test in this experiment, one probe of the probe station was first connected to the TiN of circular electrode point with the radius of 55 μm, and the other probe was connected to the rectangular electrode of TiN (5500×4000 μm^2) to form measuring circuit, as shown in Fig. S1(b). The capacitance of the MIS structure capacitor was equivalent to the series connection of two capacitors C_1 and C_2. Their relationship was described by the following equation:

$$\frac{1}{C}=\frac{1}{C_1}+\frac{1}{C_2} \tag{S1}$$

Where C was the total capacitance, C_1 and C_2 were the two capacitors of the series circuit respectively.

In addition, the formula for the plate capacitor was described as:

$$C=\frac{\varepsilon_0\varepsilon_r S}{d} \tag{S2}$$

Where C was the total capacitance, ε_0 was the vacuum dielectric constant, ε_r was the dielectric constant of the film, S was the area of the capacitor plate, d was the plate spacing of the capacitor.

According to the above two formulas S1 and S2, the formula of the plate capacitor can also be described as:

$$C = \frac{C_1 C_2}{C_1 + C_2} = \frac{\varepsilon_0 \varepsilon_r S_1 S_2}{d(S_1 + S_2)} \qquad \text{(S3)}$$

Since S_1 ? S_2, the result of the total capacitance measurement (C) was similar to the electrical performance of an order of magnitude smaller capacitor (C_2).

In addition, the calculation formula for the capacitance of the plate capacitor can also be expressed as:

$$C = \frac{Q}{U} = \frac{P \times S}{U} \qquad \text{(S4)}$$

Where C was the total capacitance, P was the polarization, S was the area of the capacitor plate, U was the applied voltage during the test.

According to the above two formulas S2 and S4, the formula of the polarization can also be described as:

$$P = \varepsilon_0 \varepsilon_{\mathrm{r}} \mathrm{E} \qquad \text{(S5)}$$

Where P was the polarization, ε_0 was the vacuum dielectric constant, ε_r was the relative permittivity of the film, E was the electric field.

Therefore, the relative permittivity (ε_r) of the film can be obtained by calculating the slope of the polarization-electric field (P-E) curve of the film sample.

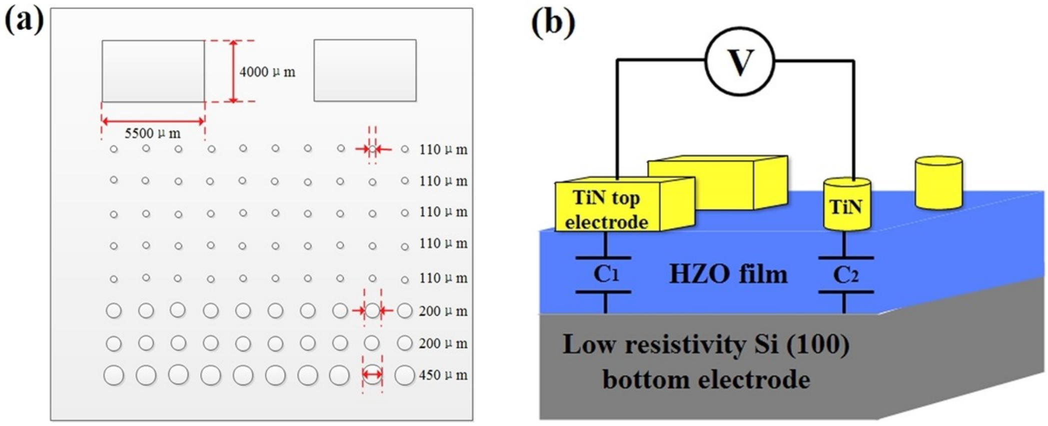

Fig. S1. (a) The hard mask design drawing of TiN top electrode. (b) Schematic diagram of MIS structure capacitor.