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Determining the Polarization Fraction of Thin Film Ferroelectric HZO with STEM EBIC

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Commonly employed as a high-*k* dielectric in the semiconductor industry, thin film hafnia (HfO_2) was discovered to exhibit ferroelectricity in 2011 [1]. Hafnia consequently became the leading candidate material for ferroelectric random access memory (FRAM) [2], and much effort has been devoted to better understanding the origins, characteristics, and optimization of its ferroelectricity. The orthorhombic $Pca2_1$ phase is generally thought to be responsible for hafnia's ferroelectricity [3, 4], but this identification is not entirely certain because of the structural similarities between the various phases present in polycrystalline hafnia films [5, 6]. Here we use scanning transmission electron microscopy (STEM) and electron beam-induced current (EBIC) imaging to study the ferroelectric properties of hafnium-zirconium-oxide (HZO).

We fabricate a metal-insulator-metal (MIM) capacitor on an electron-transparent Si_3N_4 membrane using optical lithography (Fig. 1A). The MIM capacitor consists of TaN/HZO/TaN (30/20/30 nm) deposited via DC sputtering and plasma-enhanced atomic layer deposition (PEALD) for the TaN and HZO, respectively [7]. A rapid thermal anneal (RTA) at 700 ˚C crystallizes the conformal HZO film. We switch the thin film HZO *in situ* with a DC voltage source attached to the bottom electrode. After the HZO film is polarized, the voltage is removed and a transimpedance amplifier attached to the top electrode collects the EBIC.

Traditional STEM is insensitive to a material's electronic properties, but EBIC can reveal local electric fields and conductivity changes [8]. Standard STEM imaging of these devices (Figs. 1 B and C) reveals little to no contrast variation within the capacitor because of the large column density where the electrodes overlap (Hf and Ta have atomic number Z of 72 and 73, respectively). Despite the device's opacity to normal STEM imaging, EBIC imaging clearly visualizes HZO grains with remanent polarization (Figs. 1 D – F). The EBIC images are acquired with no electric field applied, so the HZO's remanent polarization is responsible for the observed EBIC. After a −5 V bias (Fig. 1D), most of the HZO grains in the EBIC image have bright contrast, indicating a persistent electric field $\boldsymbol{E}$ pointing from the bottom electrode (BE) to the top electrode (TE). Applying a +5 V bias changes the EBIC contrast in much of the MIM capacitor from bright to dark (Fig. 1E), indicating that the polarization has reversed. The difference image (Fig. 1F) highlights the portions of the MIM capacitor that switch with a ±5 V applied voltage. About 60% of the MIM capacitor is switchable under our test conditions, and the most active region is located on the right side of the capacitor.

Our observations demonstrate EBIC imaging's capability to characterize thin film ferroelectrics inside the TEM. However, because of the electrode material's opacity, characterizing the HZO's crystal

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structure is difficult. Future studies will involve samples with different electrode materials and/or geometries to better correlate HZO's electronic and crystal structures [9].

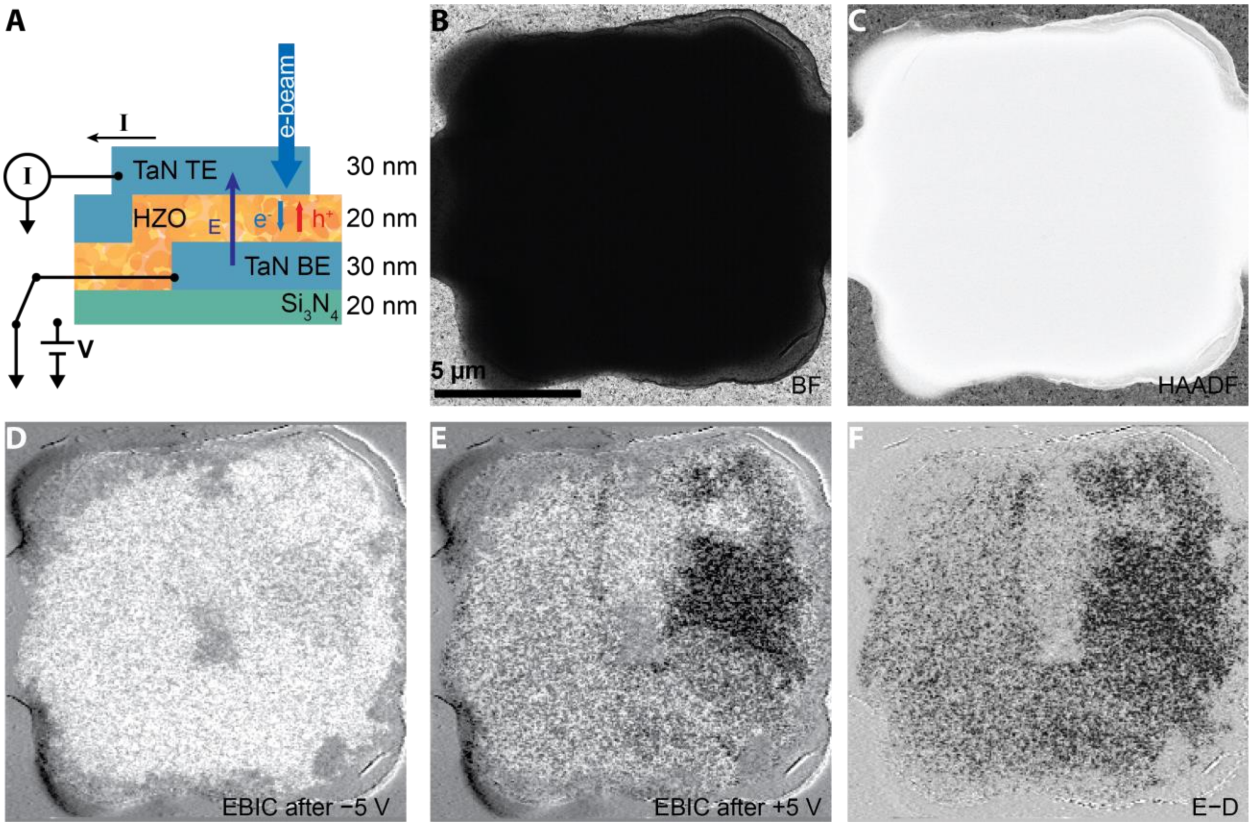

Figure 1. Schematic of device cross-section (A), BF STEM image (B), HAADF STEM image (C), and STEM EBIC images (D – F) of a TaN/HZO/TaN capacitor. The regular STEM images (B and C) show little to no contrast variation within the capacitor, but the HZO outside the capacitor is clearly crystalline. Bright EBIC contrast after a – 5 V bias (D) flips to dark contrast after a + 5 V bias (E), as highlighted by the difference image (F = E – D).

References:

[1] TS Böscke et al., Applied Physics Letters **99** (2011), 102903.
[2] J Müller et al., ECS Journal of Solid State Science and Technology **4** (2015), p. N30.
[3] X Sang et al., Applied Physics Letters **106** (2015), p. 162905.
[4] T Mikolajick et al., MRS Bulletin **43** (2018), p. 340.
[5] MH Park et al., Advanced Materials **27** (2015), p. 1811.
[6] R Batra et al., The Journal of Physical Chemistry C **121** (2017), p. 4139.
[7] SS Fields et al., Applied Physics Letters **118** (2021), 102901.
[8] WA Hubbard, Advanced Functional Materials **32** (2021), p. 2102313.
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