Supplementary Materials

Ferroelectricity in Si-Doped Hafnia: Probing Challenges in Absence of Screening Charges

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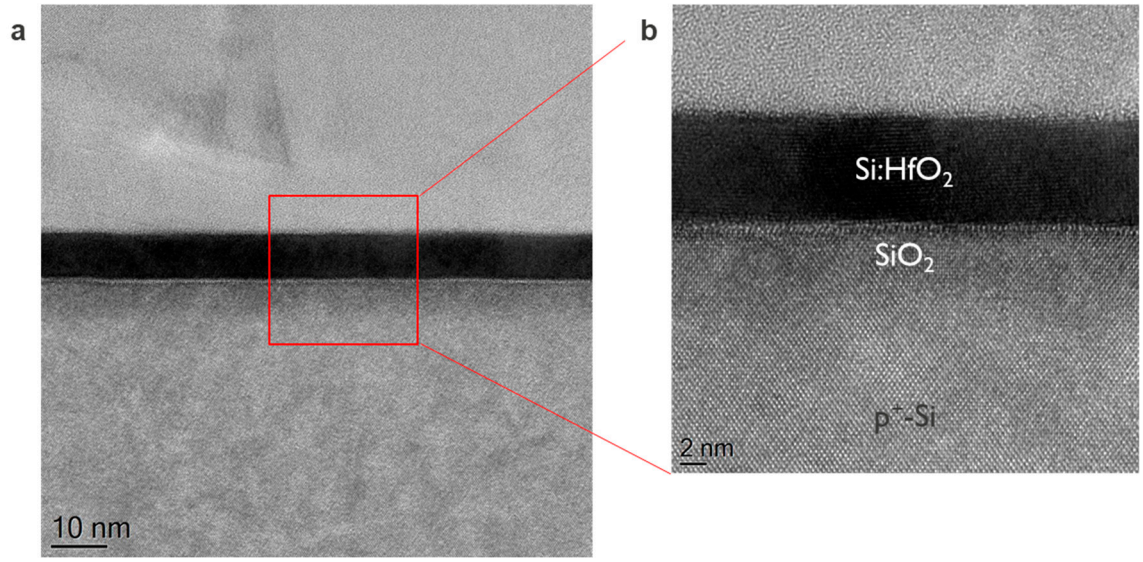

Figure S1 Si-doped HfO_2 measured by high resolution transmission electron microscopy (HRTEM), the presence of sharp interfaces (**a**) and the polycrystalline nature of the oxide layer (**b**) are both visible.

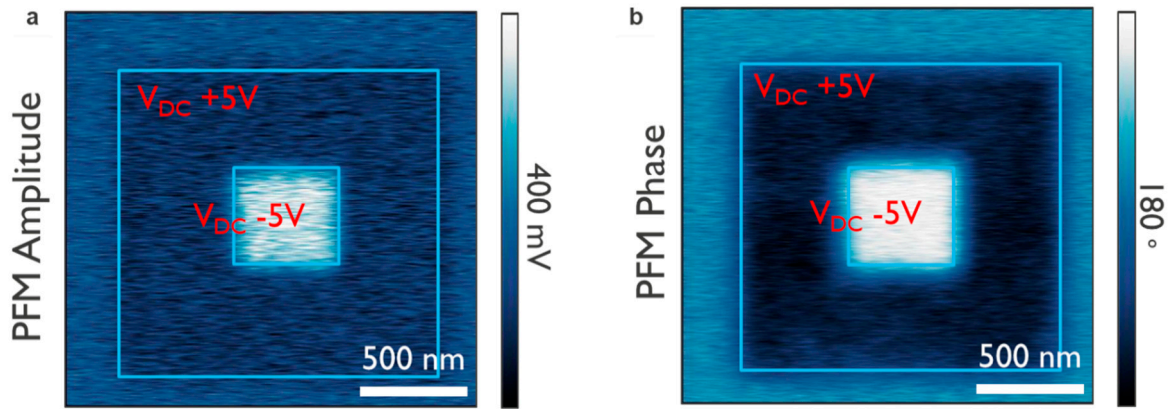

Figure S2 Conventional non-resonant PFM images are shown in figure, (**a**) amplitude and (**b**) phase contrast. The probed area has been previously polarized using two scans with a dc bias applied between tip and sample as indicated in highlighted regions. It is noteworthy that independently from the percentage of orthorhombic phase contained in the sample, the PFM response results always as a complete and uniform readout clearly suggesting a undesired surface charging-induced PFM contrast.

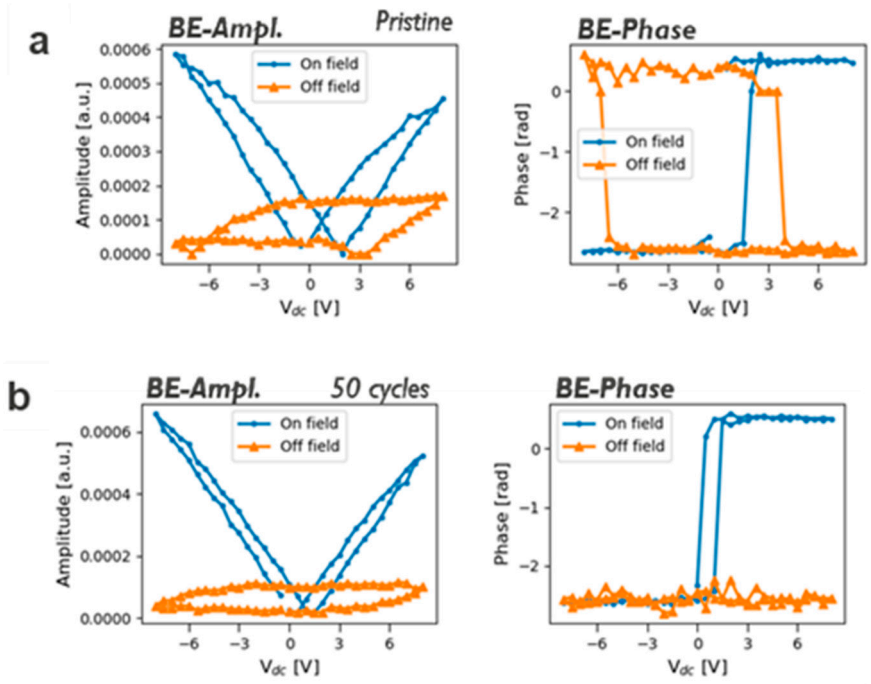

Figure S3 BE-PFM point-spectroscopy study, comparison between pristine (**a**) and cycled (**b**) Si:HfO_2. Amplitude and phase readout are presented for the same location that is stressed with multiple dc I-V sweeps with consequent collapse of the BE-PFM response.

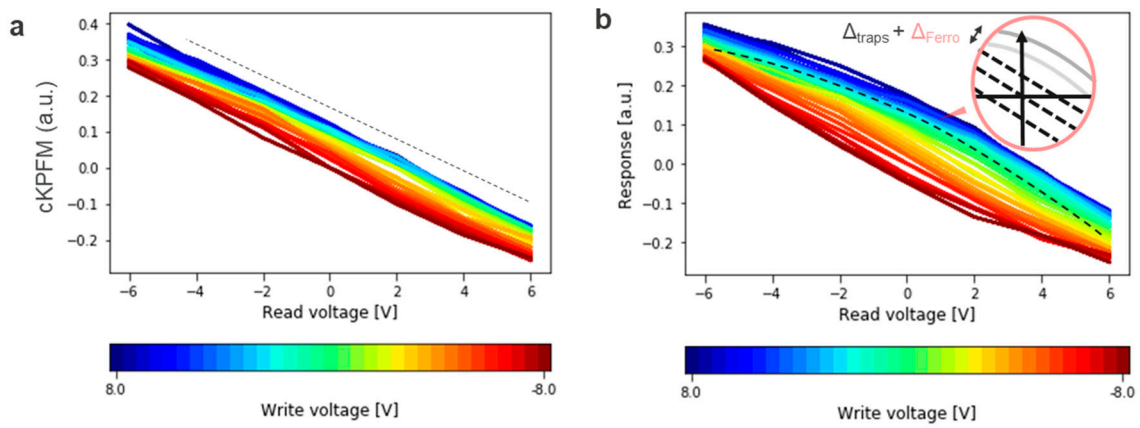

Figure S4 The case of cKPFM for a pure dielectric layer (undoped-HfO_2) is presented in (a) as a comparison for the results obtained in the Si-doped HfO_2. The two layers have the same thickness (8 nm) and the appearance of a linear dependence in the cKPFM readout suggest a purely electrostatic behavior for the undoped layer (as expected) with clear differences for the case of doped layers.

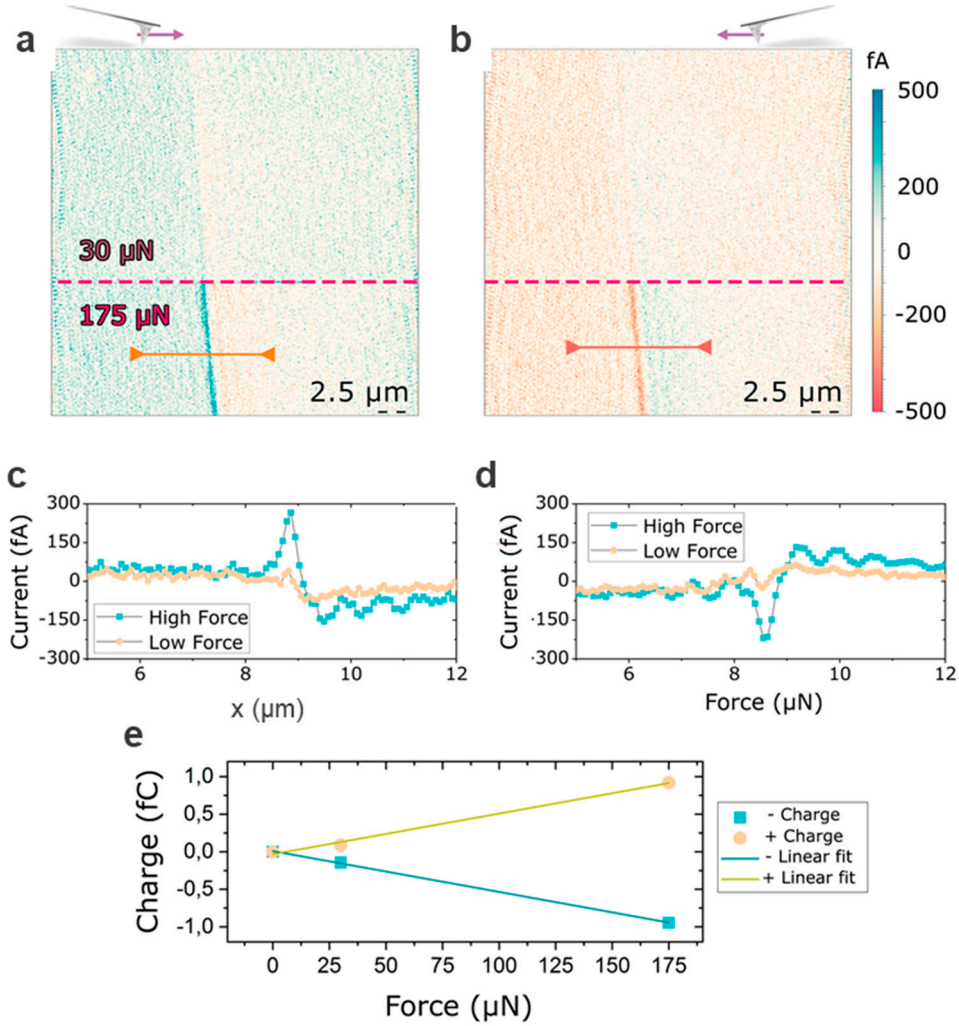

Figure S5 DPFM trace (**a**) and retrace (**b**) for PPLN test structure probes with two distinctive load forces. (**c**) PPLN extracted profiles at the domain walls as a function of the load forces, clearly indicating a pressure dependence between charge generated and force. (**d**) PPLN extracted profiles at the domain walls (retrace). (**e**) The integration of the line profiles is sued to generate a plot that relates charge as a function of applied force that shows a linear dependence for PPLN.