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### Authors

Cheema, Suraj S  
Shanker, Nirmaan  
Wang, Li-Chen  
[et al.](#)

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# 1 Ultrathin ferroic HfO<sub>2</sub>-ZrO<sub>2</sub> superlattice gate stack for advanced transistors

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3 Suraj S. Cheema,<sup>1,\*†</sup> Nirmaan Shanker,<sup>2\*</sup> Li-Chen Wang,<sup>1</sup> Cheng-Hsiang Hsu,<sup>2</sup> Shang-Lin Hsu,<sup>2</sup>  
4 Yu-Hung Liao,<sup>2</sup> Matthew San Jose,<sup>3</sup> Jorge Gomez,<sup>3</sup> Wriddhi Chakraborty,<sup>3</sup> Wenshen Li,<sup>2</sup> Jong-Ho  
5 Bae,<sup>2</sup> Steve K. Volkman,<sup>4</sup> Daewoong Kwon,<sup>2</sup> Yoonsoo Rho,<sup>5</sup> Gianni Pinelli,<sup>6</sup> Ravi Rastogi,<sup>6</sup> Do  
6 minick Pipitone,<sup>6</sup> Corey Stull,<sup>6</sup> Matthew Cook,<sup>6</sup> Brian Tyrrell,<sup>6</sup> Vladimir A. Stoica,<sup>7</sup> Zhan Zhang,<sup>8</sup>  
7 John W. Freeland,<sup>8</sup> Christopher J. Tassone,<sup>9</sup> Apurva Mehta,<sup>9</sup> Ghazal Soheli,<sup>10</sup> David Thompson,<sup>10</sup>  
8 Dong Ik Suh,<sup>11</sup> Won-Tae Koo,<sup>11</sup> Kab-Jin Nam,<sup>12</sup> Dong Jin Jung,<sup>12</sup> Woo-Bin Song,<sup>12</sup> Chung-Hsun  
9 Lin,<sup>13</sup> Seunggeol Nam,<sup>14</sup> Jinseong Heo,<sup>14</sup> Narendra Pariha,<sup>15</sup> Costas P. Grigoropoulos,<sup>5</sup> Padraic  
10 Shafer,<sup>16</sup> Patrick Fay,<sup>3</sup> Ramamoorthy Ramesh,<sup>1,17,18</sup> Souvik Mahapatra,<sup>15</sup> Jim Ciston,<sup>19</sup> Suman  
11 Datta,<sup>3</sup> Mohamed Mohamed,<sup>6</sup> Chenming Hu,<sup>2</sup> Sayeef Salahuddin<sup>2,18†</sup>

12 <sup>1</sup>*Department of Materials Science and Engineering, University of California, Berkeley, CA, USA*

13 <sup>2</sup>*Department of Electrical Engineering and Computer Sciences, University of California, Berkeley, CA,*  
14 *USA*

15 <sup>3</sup>*Department of Electrical Engineering, University of Notre Dame, Notre Dame, IN, USA*

16 <sup>4</sup>*Applied Science & Technology, University of California, Berkeley, CA, USA*

17 <sup>5</sup>*Laser Thermal Laboratory, Department of Mechanical Engineering, University of California,*  
18 *Berkeley, CA, USA*

19 <sup>6</sup>*Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, MA, USA*

20 <sup>7</sup>*Department of Materials Science and Engineering, Pennsylvania State University, University Park, PA,*  
21 *USA*

22 <sup>8</sup>*Advanced Photon Source, Argonne National Laboratory, Lemont, IL, USA*

23 <sup>9</sup>*Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory, Menlo Park,*  
24 *CA, USA*

25 <sup>10</sup>*Applied Materials, Santa Clara, CA, USA*

26 <sup>11</sup>*SK Hynix Inc., Icheon, Gyeonggi-do 17336, Korea*

27 <sup>12</sup>*Semiconductor R&D Center, Samsung Electronics, Gyeonggi-do, 445-330, Korea*

28 <sup>13</sup>*Logic Technology Development, Intel Corporation, Hillsboro, Oregon 97124, USA*

29 <sup>14</sup>*Samsung Advanced Institute of Technology, Samsung Electronics, Gyeonggi-do, 445-330, Korea*

30 <sup>15</sup>*Department of Electrical Engineering, Indian Institute of Technology, Bombay, Mumbai 400076, India*

31 <sup>16</sup>*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA, USA*

32 <sup>17</sup>*Department of Physics, University of California, Berkeley, CA, USA*

33 <sup>18</sup>*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA, USA*

34 <sup>19</sup>*National Center for Electron Microscopy, Molecular Foundry, Lawrence Berkeley National Laboratory,*  
35 *Berkeley, CA, USA*

36 **\*These authors contributed equally to this work**

37 **† Correspondence to: [s.cheema@berkeley.edu](mailto:s.cheema@berkeley.edu) (S.S.C), [sayeef@berkeley.edu](mailto:sayeef@berkeley.edu) (S.S.)**

38

39 With the scaling of lateral dimensions in advanced transistors, an increased gate capacitance  
40 is desirable both to retain the control of the gate electrode over the channel and to reduce  
41 the operating voltage<sup>1</sup>. This led to the adoption of high- $\kappa$  dielectric HfO<sub>2</sub> in the gate stack in  
42 2008<sup>2</sup>, which remains as the material of choice to date. Here, we report HfO<sub>2</sub>-ZrO<sub>2</sub>  
43 superlattice heterostructures as a gate stack, stabilized with mixed ferroelectric-  
44 antiferroelectric order, directly integrated onto Si transistors, and scaled down to  $\sim 20$  Å,  
45 the same gate oxide thickness required for high performance transistors. The overall EOT  
46 (equivalent oxide thickness) in metal-oxide-semiconductor capacitors is equivalent to  $\sim 6.5$   
47 Å effective SiO<sub>2</sub> thickness. Such a low effective oxide thickness and the resulting large  
48 capacitance cannot be achieved in conventional HfO<sub>2</sub>-based high- $\kappa$  dielectric gate stacks  
49 without scavenging the interfacial SiO<sub>2</sub>, which has adverse effects on the electron transport  
50 and gate leakage current<sup>3</sup>. Accordingly, our gate stacks, which do not require such  
51 scavenging, provide substantially lower leakage current and no mobility degradation. This  
52 work demonstrates that ultrathin ferroic HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers, stabilized with competing  
53 ferroelectric-antiferroelectric order in the 2 nm thickness regime, provide a new path  
54 towards advanced gate oxide stacks in electronic devices beyond the conventional HfO<sub>2</sub>-  
55 based high- $\kappa$  dielectrics.  
56

57 With the two-dimensional scaling of silicon field-effect transistors reaching fundamental limits<sup>1</sup>,  
58 new functional improvements to transistors<sup>4</sup>, as well as novel computing paradigms and vertical  
59 device integration at the architecture-level<sup>5</sup>, are currently under intense investigation<sup>1,4,6</sup>. Gate  
60 oxides play a critical role in this endeavor as a common performance booster for all devices,  
61 including silicon<sup>2</sup>, new high-performance channel materials<sup>7,8</sup>, and even materials suitable for  
62 three-dimensional integrated transistors<sup>9,10</sup>. Indeed, the gate oxide transition from SiO<sub>2</sub> to high- $\kappa$   
63 dielectric (DE) is considered a paradigm shift in computing technology. In this context,  
64 ferroelectric oxides offer new functionalities<sup>11</sup> considered promising for energy-efficient  
65 electronics<sup>4,9</sup>. The advent of atomic layer deposition (ALD) grown ferroelectric doped-HfO<sub>2</sub><sup>12</sup>  
66 has overcome much of the material compatibility issues that plague traditional perovskite-based  
67 ferroelectric materials<sup>2</sup>. In addition, ferroic order persists down to a thickness of 1 nm in this  
68 system<sup>13-15</sup>, fostering integration into the most aggressively-scaled devices in which the state-of-  
69 the-art high- $\kappa$  oxide thickness is less than 2 nm.

70  
71 In an advanced silicon transistor, the gate oxide is a combination of two distinct layers. The first  
72 is an interfacial SiO<sub>2</sub> formed with a self-limiting process, resulting in  $\sim$ 8.0-8.5 Å thickness<sup>16</sup>.  
73 The next is the high- $\kappa$  (HK) dielectric HfO<sub>2</sub> layer that is typically  $\sim$ 2 nm in thickness. Higher  
74 capacitance of this series combination is desirable to suppress short channel effects. The  
75 capacitance is conventionally represented by equivalent oxide thickness (EOT),  $EOT = t_{SiO_2} +$   
76  $t_{HK}/(\epsilon_{HK}/\epsilon_{SiO_2})$ , where lower EOT represents higher capacitance. Therefore, the EOT minimum  
77 value is limited by the interfacial SiO<sub>2</sub> thickness. Typically, with HfO<sub>2</sub> as the high- $\kappa$  layer, the  
78 EOT is approximately 9.5 Å. To go below this value<sup>17,18</sup>, the semiconductor industry has  
79 implemented sophisticated scavenging techniques<sup>16,18,19</sup> to reduce the SiO<sub>2</sub> thickness after the  
80 full gate stack has been deposited. Although this technique is effective in scaling EOT, the  
81 thinner SiO<sub>2</sub> results in undesirable leakage<sup>20</sup>, mobility degradation<sup>2,16</sup> and reliability issues.

82  
83 In this work, we present an ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub> superlattice gate stack that exploits mixed  
84 ferroelectric-antiferroelectric (FE-AFE) order (Fig. 1a,b), stabilized down to 2 nm thickness –  
85 the same high- $\kappa$  oxide thickness used in advanced transistors. When integrated on silicon, the  
86 gate stack shows an overall EOT of 6.5 Å, even though both transmission electron microscopy  
87 (TEM) and electrical characterization reveal an 8.0-8.5 Å interfacial SiO<sub>2</sub> thickness, as is  
88 typically expected from a chemically grown interfacial layer without scavenging. No scavenging  
89 of the interfacial SiO<sub>2</sub> results in substantially lower leakage current for the same EOT compared  
90 to benchmarks established by major semiconductor industries<sup>3</sup>. In addition, no mobility  
91 degradation is observed as EOT is scaled with these HfO<sub>2</sub>-ZrO<sub>2</sub> ferroic gate stacks. Therefore,  
92 ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub> gate stacks exploiting ferroic order offer a promising pathway toward  
93 advanced energy-efficient transistors.

## 94 95 **Ultrathin FE-AFE HfO<sub>2</sub>-ZrO<sub>2</sub> superlattices**

96  
97 Thin films of HfO<sub>2</sub>-ZrO<sub>2</sub> are grown using ALD in which the nanolaminate periodicity is dictated  
98 by the sequence of Hf:Zr (4:12) ALD cycles before the Hf-Zr superstructure is repeated various  
99 times (Fig. 1c, Methods). After top metal deposition, the entire gate stack undergoes a low-  
100 temperature post-metal anneal (200 C, 60s, N<sub>2</sub>) which does not interfere with the HfO<sub>2</sub>-ZrO<sub>2</sub>  
101 multilayer structure: various characterization techniques – synchrotron x-ray reflectivity (XRR),  
102 layer-resolved electron energy loss spectroscopy (EELS) and angle-resolved X-ray photoelectric

103 spectroscopy (XPS) – confirm the expected Hf 4 Å - Zr 12 Å periodicity (Extended Data Fig. 1).  
104 The underlying mixed ferroic order in these HfO<sub>2</sub>-ZrO<sub>2</sub> heterostructures is structurally  
105 established by high-resolution transmission electron microscopy (TEM) (Fig. 1d, Extended Data  
106 Fig. 2e,f) and in-plane grazing incidence diffraction (Fig. 1e, Extended Data Fig. 2a). Both  
107 techniques indicate the presence of the tetragonal (P4<sub>2</sub>/nmc, *T*-) and orthorhombic (Pca2<sub>1</sub>, *O*-)  
108 phase, which correspond to antiferroelectric and ferroelectric order in fluorite-structure films,  
109 respectively. Furthermore, local TEM imaging indicates the ferroelectric (orthorhombic) and  
110 antiferroelectric (tetragonal) phases are laterally intertwined (Fig. 1d, Extended Data Fig. 2e,f).  
111 Synchrotron X-ray spectroscopy and optical spectroscopy further confirm the presence of  
112 inversion symmetry breaking in the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> (HZH) multilayer (Extended Data  
113 Fig. 2c,d).

114  
115 We note here that the original Kittel view of an "antipolar" crystal structure<sup>21</sup> does not apply to  
116 the nonpolar tetragonal lattice attributed to fluorite-structure antiferroelectricity. Instead, the  
117 field-induced tetragonal-to-orthorhombic (nonpolar-to-polar) phase interconversion as the origin  
118 of antiferroelectricity has been examined in both ZrO<sub>2</sub><sup>22,23</sup> and HfO<sub>2</sub><sup>22,24</sup>. Therefore, at low  
119 electric fields, the mixed ferroelectric-antiferroelectric behavior is analogous to a ferroelectric-  
120 dielectric (polar-nonpolar) heterostructure, which can impart depolarization fields on the  
121 ferroelectric layer<sup>25</sup>. The laterally-intertwined nonpolar-polar phases present in the ultrathin  
122 HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructure are conducive to flattening the ferroelectric energy landscape  
123 through the aforementioned depolarization fields<sup>26-28</sup> (Fig. 1a). Furthermore, heterogeneous  
124 elastic energies in structurally inhomogeneous systems have been shown to destabilize long-  
125 range polarization, suppress polarization, and thereby flatten energy landscapes<sup>28</sup>.

126  
127 Additionally, the polarization in the ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> multilayer exhibit an in-plane  
128 component. 2D reciprocal space maps indicate a strong out-of-plane (111) texture (Extended  
129 Data Fig. 2b), which is consistent with TEM images demonstrating vertically-stacked planes of  
130 111- interplanar lattice spacing (Extended Data Fig. 2f). Therefore, considering the polarization  
131 is directed along a principal lattice direction for the Pca2<sub>1</sub> orthorhombic structure, the highly-  
132 oriented out-of-plane (111) texture indicates an in-plane projected polarization. The in-plane  
133 polarization introduces additional depolarization field due to the electrostatic coupling with the  
134 nonpolar antiferroelectric phases in the lateral direction. Notably, exploiting inhomogeneity to  
135 induce depolarization fields and enhance susceptibility has been demonstrated for perovskites  
136 exhibiting heterogeneous polar-nonpolar regions<sup>28</sup>. Following the same underlying mechanisms,  
137 our work demonstrates that it is possible to stabilize a mixed nonpolar-polar phase competition in  
138 2-nm-thick binary oxide films and enhance its permittivity. We also note that flattening of the  
139 energy landscape via depolarization fields is the same underlying principle of the negative  
140 capacitance (NC) effect<sup>11,29</sup>, in which depolarization fields stabilize the ferroelectric locally at a  
141 higher energy state compared to the ground state of an isolated, homogeneous ferroelectric,  
142 leading to negative curvature energy landscapes<sup>30,31</sup>.

143  
144 To confirm the higher susceptibility in the mixed AFE-FE system directly, we have performed  
145 capacitance-voltage (*C-V*) hysteresis loops in metal-insulator-metal (MIM) capacitor structures  
146 on thicker films with the same superlattice periodicity (Fig. 2a). Besides features indicative of  
147 mixed FE-AFE order, the total capacitance for the superlattice is larger than both conventional  
148 AFE ZrO<sub>2</sub> and FE Zr:HfO<sub>2</sub> of the same thickness (Fig. 2a), demonstrating enhanced

149 susceptibility. To quantify the permittivity, capacitance measurements were performed across the  
150 superlattice thickness series. These measurements yield an extracted permittivity of  $\sim 52$  (Fig.  
151 2b, Methods), which is larger than both FE orthorhombic Zr:HfO<sub>2</sub> and AFE tetragonal ZrO<sub>2</sub>  
152 values<sup>32</sup>.

153  
154 To further understand the ferroic evolution in these HfO<sub>2</sub>-ZrO<sub>2</sub> superlattices, we performed low  
155 temperature measurements where enhanced FE phase stabilization is expected. Indeed,  
156 temperature-dependent *C-V* loops for thicker HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers demonstrate an evolution  
157 from mixed-ferroic to FE-like hysteresis upon cooling slightly below room temperature ( $\sim 240$   
158 K, Fig. 2c), consistent with temperature-dependent X-ray spectroscopy indicating transition from  
159 mixed tetragonal-orthorhombic phase to predominately orthorhombic structure at similar  
160 temperatures (Extended Data Fig. 3c). The capacitance decrease upon cooling as the system  
161 moves away from the highly-susceptible mixed ferroic phase is consistent with previous work on  
162 NC in FE-DE systems<sup>29</sup> which establishes the energy landscape link between enhanced  
163 capacitance and susceptibility near phase transitions. Notably, the intertwined FE-AFE phases  
164 within the superlattice and resulting enhancement in susceptibility from the competition of FE  
165 and AFE phases is analogous to negative stiffness composites of ferroelastics within a metal  
166 matrix<sup>33,34</sup>, i.e., the mechanical analog to NC.

#### 167 168 **Ultrathin FE-AFE HfO<sub>2</sub>-ZrO<sub>2</sub> MOS capacitors**

169  
170 Next, the superlattices were grown on Si substrates in metal-oxide-semiconductor (MOS)  
171 capacitor structures. A self-limiting chemical oxide SiO<sub>2</sub> was grown first, resulting in  $\sim 8.0$ - $8.5$  Å  
172 thickness<sup>3</sup>, following the standard practice in advanced Si devices (Methods). Subsequently, a  
173 20-cycle thick multilayer was grown with ALD following the same stacking as before i.e.  
174 Hf:Zr:Hf 4:12:4. Accumulation *C-V* curves of the superlattice stack results in significantly larger  
175 capacitance in comparison to other conventional stacks – DE HfO<sub>2</sub>, AFE ZrO<sub>2</sub>, FE Zr:HfO<sub>2</sub> – of  
176 the same 20 Å thickness (Fig. 2d). Furthermore, the Hf:Zr:Hf 4:12:4 trilayer demonstrates  
177 enhanced capacitance compared to a bilayer (Hf:Zr 8:12) and solid solution (Hf:Zr [2:3]<sub>4</sub>) of the  
178 same thickness and Hf:Zr composition (Fig. 2e).

179  
180 Notably, the composition in our films is close to where several previous reports have postulated a  
181 possible morphotropic phase boundary (MPB) in thicker HfO<sub>2</sub>-ZrO<sub>2</sub> solid solution films<sup>35</sup>. Note  
182 MPB systems follows strict symmetry requirements<sup>36</sup>, which have not been established for the  
183 HfO<sub>2</sub>-ZrO<sub>2</sub> system. In our ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> multilayers, the negative free energy  
184 curvature of the polar FE O-phase compensates the positive curvature of the nonpolar AFE T-  
185 phase (Fig. 1a), leading to a flattened energy landscape. Similarly, energy landscape flattening is  
186 postulated as the thermodynamic origin of enhanced piezoelectric response in the canonical  
187 perovskite ferroelectrics<sup>36</sup>, in which multiple crystal symmetries are nearly degenerate across a  
188 composition phase boundary (MPB). However, a critical distinction is that here, the overall  
189 energy landscape flattening, and corresponding increase in capacitance, is determined by the  
190 stacking of the atomic-scale HfO<sub>2</sub>-ZrO<sub>2</sub> layers, and not the volume fraction of the constituent  
191 elements<sup>37</sup>: solid solution of the same Hf:Zr composition does not provide the same high  
192 capacitance (Fig. 2e). Furthermore, compared to HfO<sub>2</sub>-ZrO<sub>2</sub> solid solutions across a range of  
193 typically-reported Zr-rich "MPB"-like compositions<sup>35</sup>, the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> multilayer  
194 demonstrates larger capacitance (Extended Data Fig. 4). This indicates the enhanced capacitance

195 in HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> films is not simply driven by doping<sup>32,38</sup>, but can instead be tuned by the  
196 configuration of the multilayer structure (Extended Data Fig. 4, 5). In the ultrathin regime,  
197 surface energies become a more dominant consideration for determining polymorphic phase  
198 stability<sup>22</sup>; accordingly, the importance of stacking is amplified.

199  
200 To quantify the observed capacitance, we have performed EOT simulations of MOS capacitors  
201 using the industry standard Synopsys simulation platform (Methods). The Hf:Zr:Hf 4:12:4  
202 trilayer stacks vary between 6.5-7.0 Å EOT (Fig. 2f), consistent over many measured capacitors.  
203 Notably, this EOT is smaller than the expected thickness of the interfacial SiO<sub>2</sub> layer (8.0-8.5 Å),  
204 as mentioned. To investigate further, high-resolution TEM of the gate stacks (Extended Data Fig.  
205 6) illustrates the SiO<sub>2</sub> thickness is indeed ~ 8.5 Å. To supplement this physical characterization,  
206 we next implemented electrical characterization of the interfacial layer via inverse capacitance vs  
207 thickness analysis of conventional dielectric HfO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> thickness series grown on the same  
208 SiO<sub>2</sub> (Methods, Extended Data Fig. 6). All thermal processing is kept the same as the HfO<sub>2</sub>-ZrO<sub>2</sub>  
209 superlattice gate stack. The extracted HfO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> permittivity – 19 and 9, respectively – is  
210 consistent with the typical dielectric phases of these two materials. Therefore, one can reliably  
211 extract the SiO<sub>2</sub> layer thickness, yielding 8 Å (Extended Data Fig. 6), consistent with the HR-  
212 TEM results and values established by the semiconductor industry<sup>3</sup>.

213  
214 Moreover, the consistent interlayer thickness extracted from both material systems indicates that  
215 neither Hf nor Al encroaches into the interfacial SiO<sub>2</sub>, which would reduce its thickness and/or  
216 increase its permittivity. This is expected considering the gate oxides are processed at much  
217 lower temperature than that needed for silicate formation<sup>39</sup> and works reporting an increased  
218 SiO<sub>2</sub> interlayer permittivity<sup>40</sup>. Furthermore, XRR, EELS and XPS data indicate that for both the  
219 undoped control HfO<sub>2</sub> gate stack and the superlattice gate stack, the HfO<sub>2</sub> layer sits right on top  
220 of SiO<sub>2</sub>, leading to the same interface in both cases (Extended Data Fig. 1). Therefore,  
221 considering the interfacial layer thickness as 8 Å, the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> multilayer gate stack  
222 demonstrates an overall EOT ~ 1.5 Å lower than the constituent SiO<sub>2</sub> thickness. We note for  
223 simplicity, we have used an EOT to quantify the capacitance of the superlattice stack; however,  
224 for a rigorous description, one should solve for the non-linearities that are expected to emerge  
225 from the ferroic nature of the gate-oxide<sup>41</sup>.

226  
227 To supplement the *C-V* evidence of capacitance enhancement, pulsed current-voltage (*I-V*)  
228 measurements of MOS capacitors integrating the ~ 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> gate stack – which  
229 can quantify the amount of charge as a function of voltage<sup>26</sup> (Methods, Extended Data Fig. 7) –  
230 demonstrate larger stored charge than if just interfacial SiO<sub>2</sub> was sitting on top of Si. This  
231 provides further electrical evidence of charge enhancement in the ultrathin mixed ferroic gate  
232 stack (Extended Data Fig. 7e). Furthermore, from these measurements, the extracted  
233 polarization- electric field relationship for just the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> multilayer (Extended Data  
234 Fig. 7f) exhibits a regime of negative slope, which mathematically corresponds to negative  
235 capacitance stabilization<sup>26</sup>.

### 236 237 **Ultrathin FE-AFE HfO<sub>2</sub>-ZrO<sub>2</sub> Device Results**

238  
239 The practical implication of this capacitance enhancement can be clearly seen in Fig. 3b, which  
240 shows leakage current vs. EOT behavior. The leakage current is measured at  $V_G - V_{fb} = -1$  V,

241 where  $V_{fb}$  is the flatband voltage of the semiconductor. All other data points on this plot are taken  
242 from reported industrial gate stacks<sup>3</sup>. The leakage current for the Hf:Zr:Hf 4:12:4 stack is  
243 substantially lower at the same EOT. Note that below 9 Å, the other gate stacks need  
244 sophisticated scavenging techniques to reduce the thickness of the interfacial SiO<sub>2</sub><sup>3</sup>. On the other  
245 hand, the ferroic gate stack can achieve ~ 6.5 Å without any scavenging, resulting in the lower  
246 leakage current (Fig. 3b).

247  
248 Furthermore, the scavenging of the interfacial SiO<sub>2</sub> leads to a loss of mobility of ~20 cm<sup>2</sup>/V-s per  
249 every Å of scavenged SiO<sub>2</sub> due to an increase in remote phonon scattering<sup>3,16</sup>. To examine how  
250 the mobility evolves with EOT, we compared transistors implementing the lower-EOT HfO<sub>2</sub>-  
251 ZrO<sub>2</sub>-HfO<sub>2</sub> gate stack compared to higher-EOT conventional HfO<sub>2</sub> gate stack, both of the same  
252 physical thickness (Methods). Notably, the mobility remains essentially the same for both stacks,  
253 demonstrating there is no fundamental change in electron transport due to the mixed-ferroic  
254 multilayer gate stack compared to the standard high-κ dielectric gate stack (Extended Data Fig.  
255 8d). Furthermore, this work demonstrates no penalty in mobility below 9 Å EOT, the point  
256 where conventional high-κ gate stacks display mobility degradation due to scavenging necessary  
257 for lowering EOT (Fig. 3c, Extended Data Fig. 8d). Indeed, raw mobility extracted from long-  
258 channel transistors integrating the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> (HZH) mixed-ferroic heterostructure  
259 gate stack exceed that of industry-reported long-channel transistors integrating standard 2 nm  
260 HfO<sub>2</sub> high-κ dielectric gate stacks<sup>42</sup> at the same EOT (Fig. 3c).

261  
262 To examine how the capacitance enhancement in the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> gate stack behaves  
263 at high frequency, radio frequency (RF) measurements were performed on the same long channel  
264 ( $L_G = 1 \mu\text{m}$ ) devices (Methods, Extended Data Fig. 9) to extract device parameters up to ~800  
265 MHz for our devices (close to the cut-off frequency). Of particular interest is the  
266 transconductance ( $g_m$ ), which is proportional to the product of capacitance and electron velocity  
267 (mobility). From  $Y$ -parameter measurements one can find AC transconductance as  $Re(Y_{21}) =$   
268  $g_m + af^2$ , where  $f$  is the frequency (Methods). This yields an AC transconductance as a function of  
269 applied gate voltage ( $V_G$ ). Plotting this dependence together with DC transconductance ( $\partial I_D / \partial V_G$   
270 from DC  $I_D$ - $V_G$ ) (Fig. 3h) illustrates DC and AC transconductance are similar, with AC  
271 transconductance roughly 15% larger at the peak value. This slightly larger AC transconductance  
272 may result from the fact that certain interface traps, which affect the DC behavior, cannot  
273 respond at frequencies larger than 100 MHz, leading to better gate control. More importantly,  
274 these RF results show that the observed capacitance enhancement is not limited to the low  
275 frequency regime<sup>43,44</sup>.

276  
277 Next, shorter channel ( $L_G = 90 \text{ nm}$ ) devices, fabricated on a silicon-on-insulator (SOI) transistor  
278 with 18 nm SOI thickness, were examined. The transfer and output characteristic of a typical  
279 transistor are shown in Fig. 3e,f. Note that the threshold voltage of this device is 0.55 V, which  
280 is consistent with the work function of W used as the gate metal. Because of this, the transistors  
281 have been driven up to 1.6 V gate voltage so that an overdrive voltage ( $V_{ov} = V_G - V_T$ ) of ~1 V  
282 can be applied. It is found that at a drain voltage ( $V_D$ ) and  $V_{ov}$  of 1 V, the drain current exceeds 1  
283 mA/ $\mu\text{m}$ . Additionally, the measured extrinsic transconductance of ~1.1 mS/ $\mu\text{m}$  (Fig. 3g)  
284 corresponds to an intrinsic transconductance of ~1.75 mS/ $\mu\text{m}$  (Methods, Extended Data Fig. 10).  
285 The transconductance is substantially larger than conventional 90 nm transistors. In addition, it is  
286 larger than control devices with HfO<sub>2</sub> gate stack of the same physical thickness, demonstrating



287 the dual benefits of the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> mixed-ferroic gate stack: low EOT without adversely  
288 affecting the electron transport.

289  
290 Finally, to probe the interface quality, especially trap-induced effects<sup>45</sup> relevant for MOSFET  
291 reliability – a very crucial aspect for commercial application – we performed positive bias  
292 temperature instability (PBTI) measurements on nFET transistors (Extended Data Fig. 8e-h). The  
293 results demonstrate very similar behavior for both the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> and control HfO<sub>2</sub> stacks  
294 of the same physical thickness, and similar to those reported in literature for high-κ HfO<sub>2</sub>  
295 stacks<sup>46</sup>. This is not unexpected; reliability characteristics are predominantly determined by the  
296 interfacial oxide and its high-κ interface<sup>46</sup>; here, both stacks have the same un-scavenged SiO<sub>2</sub>  
297 interlayer (Extended Data Fig. 1). Furthermore, stress measurements on capacitors demonstrate  
298 negligible  $V_{fb}$  shift and non-existent capacitance degradation with increased stress time  
299 (Extended Data Fig. 8i,j).

## 300 301 **Discussion**

302  
303 Capacitance enhancement via negative capacitance (NC) has been demonstrated for ferroelectric-  
304 dielectric superlattices in many single-crystalline perovskite-structure systems<sup>30,31,47,48</sup>. This  
305 work demonstrates that the same enhancement is possible in HfO<sub>2</sub>-ZrO<sub>2</sub> fluorite-structure  
306 superlattices on Si, which exhibit mixed ferroelectric-antiferroelectric (polar-nonpolar) order in  
307 films as thin as just ~ 2 nm. The ability to go down to such thickness and still stabilize  
308 competing ferroic order, conducive for NC mediated capacitance enhancement, is very important  
309 for advanced electronic devices as dimensional scaling requires ultrathin gate stacks.  
310 Furthermore, this work establishes the critical role of atomic-layer stacking – as opposed to  
311 conventional doping techniques<sup>32,38</sup> – in controlling the ferroic phase space and permittivity of  
312 fluorite-structure oxides down to ultrathin limits, leveraging its unique size effects<sup>13-15</sup> and rich  
313 antiferroelectric-ferroelectric polymorphs<sup>22,23</sup>. When this mixed phase HfO<sub>2</sub>-ZrO<sub>2</sub> multilayer is  
314 integrated on Si, the gate stack exhibits a capacitance enhancement, lowering the EOT below a  
315 threshold which traditionally required careful scavenging of interfacial SiO<sub>2</sub>, which would  
316 otherwise degrade mobility<sup>3</sup>. Additionally, this EOT is achieved at over an order of magnitude  
317 lower leakage current. Therefore, harnessing atomic-scale layering in ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub> ferroic  
318 gate oxides presents a promising materials design platform for future Si transistors beyond the  
319 conventional high-κ dielectrics which have spurred semiconductor industry scaling over the past  
320 two decades.

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455  
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457 optimization of the superlattice oxide heterostructure and its ferroic characterization. N.S. and C.-H.H.  
458 performed capacitor fabrication. C.-H.H. helped optimize annealing treatments. N.S. and S.S.C.  
459 performed capacitor measurements and analysis. L.-C.W. fabricated the transistors and performed DC  
460 characterization. D.W.K. and J.B. developed the initial processes for transistor fabrication. Y.-H.L.  
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476 **Correspondence and requests for materials** should be addressed to S.S.C. (s.cheema@berkeley.edu)  
477 and S.S. (sayeef@berkeley.edu).

478 **Fig. 1. Atomic-scale design of negative capacitance in ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub>.** (a) Energy  
479 landscape flattening. A ferroelectric (FE) double-well energy landscape is flattened by the  
480 depolarization field energies (Dep) originating from electrostatic and elastic inhomogeneities  
481 present in the laterally-arranged polar-nonpolar (orthorhombic ferroelectric - tetragonal  
482 antiferroelectric) thin film system. The energy landscape flattening increases the overall system's  
483 permittivity, as susceptibility is proportional to the inverse landscape curvature; such flattening is  
484 analogous to negative capacitance stabilization<sup>29,30,49</sup>. (b) Engineering ferroic phase competition  
485 in the HfO<sub>2</sub>-ZrO<sub>2</sub> fluorite-structure system. Beyond the conventionally-studied tuning parameters  
486 – composition, electric field, temperature<sup>32,38</sup> – here we introduce dimensional confinement via  
487 superlattice layering to tailor ferroic phase competition at the atomic-scale. (c) Schematic of the  
488 HfO<sub>2</sub>-ZrO<sub>2</sub> fluorite-structure multilayer on Si; the heterostructures maintain distinct layers (i.e.  
489 not solid solution alloys) based on EELS, XRR, and depth-resolved XPS (Extended Data Fig. 1).  
490 The role of the layering on the underlying ferroic order and capacitance is studied by electrical  
491 measurements as a function of HfO<sub>2</sub>-ZrO<sub>2</sub> stacking structure and annealing temperature  
492 (Extended Data Fig. 4 and 5, respectively). (d) HR-TEM image of the atomic-scale HfO<sub>2</sub>-ZrO<sub>2</sub>-  
493 HfO<sub>2</sub> trilayer (top) and extracted *d*-lattice spacings (bottom) corresponding to the fluorite-  
494 structure AFE tetragonal (P4<sub>2</sub>/nmc, red) and FE orthorhombic (Pca2<sub>1</sub>, blue) phases, respectively.  
495 The layer delineations are approximate, as the HfO<sub>2</sub>-ZrO<sub>2</sub> and SiO<sub>2</sub> interlayer thicknesses are  
496 more rigorously determined by XRR and TEM analysis (Extended Data Fig. 1 and 6,  
497 respectively). Note imaging the crystallinity of the HfO<sub>2</sub>-ZrO<sub>2</sub> layers requires mistilt with respect  
498 to the Si lattice (Methods). (e) Synchrotron IP-GiD demonstrating the presence of both the AFE  
499 *T*-phase (101)<sub>*t*</sub> and FE *O*-phase (111)<sub>*o*</sub> reflections whose *d*-lattice spacings are consistent with  
500 those extracted from TEM. Detailed indexing for structural identification is provided by wide-  
501 angle synchrotron diffraction (Extended Data Fig. 2a).

502 **Fig. 2. Enhanced capacitance in ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub> mixed-ferroic heterostructures. (a)**  
503 **MIM *C-V* hysteresis loops for a mixed FE-AFE HfO<sub>2</sub>-ZrO<sub>2</sub> multilayer demonstrating higher**  
504 **capacitance compared against its AFE (ZrO<sub>2</sub>) and FE (Zr:HfO<sub>2</sub>) counterparts of the same**  
505 **thickness. (b) Inverse capacitance versus thickness for MIM HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers up to 5**  
506 **superlattice repeats (10 nm); the extracted permittivity of 52 is large for HfO<sub>2</sub>-based oxides. (c)**  
507 **MIM *C-V* hysteresis loops for HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers of the same periodicity demonstrating an**  
508 **evolution from mixed-ferroic to FE-like hysteresis upon cooling slightly below room**  
509 **temperature. The proximity to the temperature-dependent phase transition (Extended Data Fig. 3)**  
510 **suggests the HfO<sub>2</sub>-ZrO<sub>2</sub> heterostructures lies near its maximum electric susceptibility position,**  
511 **ideal for negative capacitance stabilization<sup>30,49</sup>. (d) MOS accumulation *C-V* of HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub>**  
512 **trilayer compared to AFE ZrO<sub>2</sub>, FE Zr:HfO<sub>2</sub>, and DE HfO<sub>2</sub>, all of the same thickness (20 Å),**  
513 **indicating mixed-ferroic behavior is optimal for enhancing capacitance. (e) Accumulation *C-V* of**  
514 **the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> (HZH) trilayer compared to bilayer and solid solutions films of the same**  
515 **thickness (ALD cycles) and composition (Hf:Zr cycles), demonstrating the capacitance**  
516 **enhancement is not simply driven by Hf:Zr composition<sup>32,38</sup>, but rather the atomic-scale stacking**  
517 **(Extended Data Fig. 4, 5). Inset: Schematic multilayer versus solid solution (Hf and Zr cations**  
518 **vertically-separated versus intermixed). (f) Accumulation *C-V* for a 2 nm HZH grown on sub-nm**  
519 **SiO<sub>2</sub> fit EOT simulations. Inset: Gate leakage of the same stack. 2 nm HZH on SiO<sub>2</sub>**  
520 **demonstrates lower EOT than the thickness of SiO<sub>2</sub> interlayer alone, carefully extracted via**  
521 **physical (8.5 Å) and electrical (8.0 Å) methodologies (Extended Data Fig. 6), providing evidence**  
522 **of capacitance enhancement via NC. Furthermore, these 2 nm ferroic gate stacks demonstrate**  
523 **amplified charge from pulsed *I-V* measurements relative to the SiO<sub>2</sub> interlayer (Extended Data**  
524 **Fig. 7), marking the thinnest demonstration of charge and/or capacitance enhancement (Extended**  
525 **Data Fig. 7).**

526 **Fig. 3. Device performance benefits utilizing ultrathin mixed-ferroic HfO<sub>2</sub>-ZrO<sub>2</sub> gate**  
527 **stacks.**

528 **(a)** EOT scaling pathways: conventional interlayer scavenging implementing standard high- $\kappa$   
529 dielectric HfO<sub>2</sub> reduces the EOT by thinning the SiO<sub>2</sub> interlayer (red), leading to leakage and  
530 mobility degradation<sup>3</sup>; integrating a ferroic NC oxide exhibiting a capacitance enhancement  
531 effect on SiO<sub>2</sub> (blue) lowers the EOT without reducing the SiO<sub>2</sub> interlayer thickness. **(b)**  
532 Leakage-EOT scaling of the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> (HZH) multilayer gate stacks (blue) benchmarked  
533 against reported HKMG literature<sup>3</sup>, including interlayer-scavenged 2 nm HfO<sub>2</sub> (red), high- $\kappa$   
534 doped HfO<sub>2</sub> (gray), and SiO<sub>2</sub>/poly-Si (black). The leakage is the lowest reported for a 6.5 Å EOT  
535 MOS capacitor on silicon<sup>3</sup>, and similar as the standard higher-EOT 2 nm HfO<sub>2</sub> high- $\kappa$  dielectric  
536 (black), due to maintaining the same SiO<sub>2</sub> thickness. **(c)** Raw mobility versus EOT for long-  
537 channel transistors integrating the 2 nm HZH (blue) versus industry-reported long-channel  
538 transistors integrating standard 2 nm HfO<sub>2</sub> (red)<sup>42</sup>, reported at 10<sup>13</sup>/cm<sup>2</sup> charge density. The raw  
539 mobility for HZH sits above the industry-reported trend line<sup>42</sup> due to scaling EOT without  
540 requiring scavenging. Mobility results are also benchmarked against other industrial HKMG  
541 reports<sup>3</sup> (Extended Data Fig. 8d). **(d)** Schematic transistor device layout which integrates the 2  
542 nm HZH gate stack. **(e, f, g)** DC  $I$ - $V$  transfer characteristics ( $I_d$ - $V_{gs}$ , e), DC output characteristics  
543 ( $I_d$ - $V_{ds}$ , f), and DC transconductance ( $g_m$ - $V_{gs}$ , g) for short-channel ( $L_G = 90$  nm) SOI transistors.  
544 Notably, the maximum on-current and  $g_m$  at  $V_{DS} = 1$  V exceeds 1 mA/ $\mu$ m and 1 mS/ $\mu$ m,  
545 respectively. **(h)** Transconductance versus gate voltage for long-channel bulk transistors ( $L_G = 1$   
546  $\mu$ m) via DC (derivative of  $I_d$ - $V_{gs}$ ) and RF ( $Re[Y_{21}]$ ) measurements at  $V_{DS} = 1$  V. Inset: De-  
547 embedded  $Re[Y_{21}]$  (open circles) as a function of squared frequency at different DC bias points  
548 extrapolated to the zero-frequency limit (dotted lines) to extract the RF  $g_m$  (Extended Data Fig.  
549 9).



## 550 **Methods**

551

### 552 **Gate stack**

553

554 **Gate oxide** Thin films of HfO<sub>2</sub>-ZrO<sub>2</sub> were grown by atomic layer deposition (ALD) in a Fiji  
555 Ultratech/Cambridge Nanotech tool (U.C. Berkeley) at 270°C in which tetrakis  
556 (ethylmethylamino) hafnium and tetrakis (ethylmethylamino) zirconium precursors are heated to  
557 75°C and water vapor is used as the oxidant. For metal-ferroelectric-insulator-semiconductor  
558 (MFIS) capacitor structures, sub-nm chemically-grown SiO<sub>2</sub> on lightly-doped Si (10<sup>15</sup> cm<sup>-3</sup>) was  
559 prepared by the standard clean (SC-1) solution (5:1:1 H<sub>2</sub>O:H<sub>2</sub>O<sub>2</sub>:NH<sub>4</sub>OH at 80°C for 10 minutes)  
560 after the Si wafer was cleaned in Piranha (120°C for 10 minutes) to remove organics and HF  
561 (50:1 H<sub>2</sub>O:HF at room temperature for 30 s) to remove any native oxide. Subsequently, HfO<sub>2</sub>-  
562 ZrO<sub>2</sub>-HfO<sub>2</sub> multilayers are deposited at 270°C by ALD. After ALD deposition, post-deposition  
563 annealing (PDA) was performed at 175°C (20 min, forming gas N<sub>2</sub>/H<sub>2</sub> background) to help cure  
564 the SiO<sub>2</sub>-oxide interface. For confirmation and reproducibility, HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> multilayers of  
565 the same ALD cycling were also deposited at MIT Lincoln Laboratory (MIT LL); after ALD  
566 deposition, PDA was performed at 250°C (1 min, N<sub>2</sub> background).

567

568 **Gate metal** For UC Berkeley capacitors, the first layer of the gate metal, TiN, is deposited by  
569 ALD (250°C, 20 cycles, 15 Å) in N<sub>2</sub> and H<sub>2</sub> plasma. Subsequently, W is deposited by sputtering  
570 (room temperature, 60 nm). For MIT LL capacitors, the gate metal, TiN, is deposited by PVD  
571 (room temperature).

572

573 **Annealing** The entire gate stack undergoes a low-temperature post-metal anneal (200°C, 1 min,  
574 N<sub>2</sub>) to cure interface defects. This low temperature does not interfere with the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub>  
575 multilayer structure, as confirmed by various characterization techniques (Extended Data Fig. 1),  
576 and maintains the mixed ferroic behavior, as high-temperature annealing would induce purely  
577 ferroelectric behavior (Extended Data Fig. 5). X-ray diffraction and TEM confirm the presence  
578 of crystalline ultrathin films despite the low deposition temperature, afforded by the low  
579 crystallization temperature of ZrO<sub>2</sub><sup>50</sup>. In fact, non-post-annealed ALD-grown ZrO<sub>2</sub> has  
580 previously demonstrated crystallization into the ferroelectric orthorhombic phase on Si<sup>51</sup>.

581

### 582 **Device fabrication**

583

584 **MOS and MIM capacitors, Bare Structures** For MOS capacitor structures, after gate stack  
585 deposition, top electrodes are defined by photolithography and dry etching. For bare structures  
586 (structural studies), the top metal is removed by chemical etching to expose the gate oxide  
587 surface. For metal-insulator-metal (MIM) capacitors, W is deposited by sputtering (room  
588 temperature, 30 nm) on a lightly-doped Si substrate as the bottom metal electrode. After ferroic  
589 film deposition by ALD, 60 nm of W is deposited by sputtering. The top electrodes are then  
590 again defined by photolithography and dry etching.

591

592 **Bulk transistors** The n-type bulk transistors were fabricated by a non-self-aligned gate-last  
593 process on bulk silicon wafers (10<sup>17</sup> cm<sup>-3</sup>) with local oxidation of silicon (LOCOS) as device  
594 isolation technique. First, a 10 nm of SiO<sub>2</sub> thermal oxide and a 30 nm of low-pressure chemical  
595 vapor deposition (LPCVD) Si<sub>3</sub>N<sub>4</sub> were grown on the Si substrates. After the active region was

596 defined by photolithography and  $\text{Si}_3\text{N}_4/\text{SiO}_2$  etching, dry oxidation was performed to form the  
597 LOCOS isolation. Next, the source/drain regions were defined by photolithography and ion  
598 implantation with an ion dose of  $3 \times 10^{15}$  ions/cm<sup>2</sup>. The dopants were then activated by a rapid  
599 thermal anneal (RTA) at 900°C for 7 min in  $\text{N}_2$  ambient. The gate stacks with the sub-nm  
600 chemically-grown  $\text{SiO}_2$ , 2 nm  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  heterostructure, and 100 nm of sputtered W gate  
601 were then deposited. After the gate fingers (from 500 nm to 50  $\mu\text{m}$ ) were patterned by  
602 photolithography and etched by inductively-coupled plasma (ICP) metal etching, the 400 nm  
603 thick interlayer dielectric (ILD)  $\text{SiO}_2$  was deposited using plasma-enhanced CVD (PECVD).  
604 Last, after the contact hole opening, the Ti/TiN contact metal was deposited by sputtering,  
605 defined by photolithography, and then etched by ICP metal etching.

607 **Short-channel SOI Transistors** The n-type short-channel transistors were fabricated by a non-  
608 self-aligned gate-last process on SOI substrates with a gate length ( $L_G$ ) down to 90 nm. First, the  
609 device layer was thinned down to 20 nm and the active regions were defined by  
610 photolithography with expose regions etched slightly into the buried oxide. The hydrogen  
611 silsequioxane (HSQ) negative resist were written by e-beam lithography as a hard mask for the  
612 ion implantation with a dose of  $5 \times 10^{15}$  ions/cm<sup>2</sup>. The dopant activation was conducted in an  
613 RTA at 900°C for 15 seconds in  $\text{N}_2$  ambient. The gate stacks with the sub-nm chemically-grown  
614  $\text{SiO}_2$ , 2 nm  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  heterostructure, 1.5 nm of PEALD TiN, and 100 nm of sputtered W  
615 were sequentially deposited. The gate region (250 nm) was then patterned by photolithography.  
616 Like the back-end process for the bulk transistors, 400 nm of ILD and sputtered Ti/TiN contact  
617 metal were deposited and defined by photolithography and ICP etching.

## 619 **Microscopy**

621 **Transmission electron microscopy** Electron microscopy was performed at the National Center  
622 for Electron Microscopy (NCEM) facility of the Molecular Foundry at Lawrence Berkeley  
623 National Laboratory (LBNL). The high-resolution bright field TEM images of  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$   
624 thin films were performed by FEI ThemIS 60-300 microscope with image aberration corrector  
625 operated at 300 kV (Fig. 1d, Extended Data Fig. 2e,f). To prepare cross-sectional TEM samples  
626 of  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  thin films, mechanical polishing was employed by using an Allied High  
627 Tech Multiprep at a 0.5° wedge to thin down the total thickness of samples down to 10  $\mu\text{m}$ .  
628 Later, Ar ion milling of the Gatan Precision Ion Milling System was utilized to make an  
629 electron-transparent sample, starting from 4 keV down to 200 eV as final cleaning energy. For  
630 high-resolution imaging, in order to capture the crystallinity of the  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  layers, the  
631 zone axis alignment required varying degrees of mistilt with respect to the Si lattice, explaining  
632 the slightly obscured Si atomic columns (Fig. 1d, Extended Data Fig. 2e,f).

634 The local interplanar  $d$ -spacing in the ultrathin  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  films (Extended Data Fig. 2e,f)  
635 was measured by DigitalMicrograph software using its line profile plus integration width  
636 analysis. For the 2 nm  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  multilayer film, the extracted interplanar lattice spacings  
637 were averaged over multiple lattice periodicities and confirmed across various local regions of  
638 the film (Extended Data Fig. 2e,f). The  $\text{SiO}_2$  interlayer thickness from low-magnification wide  
639 field-of-view (FoV) imaging was determined by the same method (Extended Data Fig. 6a). In  
640 particular, the intensity line scan from the wide FoV image (Extended Data Fig. 6a) is obtained  
641 from averaging across the entire FoV specified by the teal-colored box (~150 nm). Next, the

642 inflection points of the intensity peak were used as the criteria to set the boundaries of the SiO<sub>2</sub>  
643 interlayer (Extended Data Fig. 6a). This methodology was also utilized to determine the  
644 boundaries of the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> layers from the EELS spectrum (Extended Data Fig. 1c).  
645 Regarding the wide FoV cross-sectional TEM (Extended Data Fig. 6a), both the low atomic  
646 weight and lack of crystallinity of the SiO<sub>2</sub> layer contribute to its weak scattering (bright color),  
647 which aids in the visual delineation of the layer boundaries and the thickness extraction from the  
648 corresponding averaged intensity line scan.

649  
650 **Optical microscopy** Second harmonic generation (SHG) measurements (Extended Data Fig. 2d)  
651 were performed with a Ti:sapphire femtosecond laser (Tsunami, Spectra Physics,  $\lambda \sim 800$  nm,  
652 frequency  $\sim 80$  MHz). The linearly polarized femtosecond laser beam was focused through 50X  
653 objective lens ( $NA \sim 0.42$ ) which results in a focal spot size of 2  $\mu\text{m}$ . The generated SHG signal  
654 was collected through the same objective lens and separated from the fundamental beam by the  
655 harmonic separator. After passing through the optical bandpass filter, the SHG signals were  
656 registered to the photon multiplier tube (PMT) without a polarizer. The fundamental beam was  
657 mechanically chopped, and the signal collected by the PMT was filtered by a lock-in amplifier to  
658 reduce the background noise. For SHG spatial mapping, a two-axis piezo stage was utilized and  
659 the coordinate was synchronized with the PMT signal. The SHG intensity was obtained by  
660 averaging the mapping signals across a 100  $\mu\text{m} \times 100 \mu\text{m}$  sample area.

661  
662 **X-ray characterization**

663  
664 **X-ray reflectivity** Synchrotron X-ray reflectivity (XRR) – performed at Sector 33-BM-C  
665 beamline of the Advanced Photon Source, Argonne National Laboratory and at Beamline 2- 1 of  
666 the Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory –  
667 confirmed the thickness of HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructures (Extended Data Fig. 1b). The  
668 overall thickness of the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructures is consistent with the growth rate ( $\sim 1$   
669  $\text{\AA}/\text{cycle}$ ) of ALD-grown Zr:HfO<sub>2</sub> as demonstrated in our previous work<sup>13</sup>. Furthermore, the  
670 presence of irregularly spaced fringes in the thicker HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructures suggests  
671 the presence of well-separated HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> layers, i.e. not a solid solution. This is  
672 confirmed by XRR fitting (Extended Data Fig. 1b) performed with the python package GenX<sup>52</sup>  
673 which considers factors such as density, roughness, and thickness.

674  
675 **In-plane grazing incidence diffraction** Synchrotron in-plane grazing-incidence diffraction  
676 (GID) (Fig. 1e, Extended Data Fig. 2a) was performed at Sector 33-ID-D beamline of the  
677 Advanced Photon Source, Argonne National Laboratory. A Pilatus-II 100K Area Detector  
678 mounted on the del-arm was used to collect diffraction signal with a grazing incidence geometry.  
679 The region-of-interest on the detector was set such that the ring-like signal was fully integrated.  
680 In-plane GID was collected by sweeping the in-plane angle  $\nu$  ( $8-50^\circ$ ) with a fixed out-of-plane  
681 grazing angle  $\delta$  ( $\delta = 0.9^\circ$ ); the corrected Bragg angle ( $2\theta$ ) over which the data is plotted and  
682 indexed is determined from the relationship  $\cos(2\theta) = \cos(\nu) \cdot \cos(\delta)$  set by the geometry of the  
683 diffractometer. The X-ray source was fixed at 16 keV ( $\lambda = 0.775 \text{\AA}$ ). In-plane diffraction yields  
684 more diffraction peaks with better defined width, likely due to the preferred orientation and disc-  
685 shape domains in the film. Therefore, in-plane GID enables clear indexing to the ferroelectric  
686 orthorhombic (Pca2<sub>1</sub>) and antiferroelectric tetragonal (P4<sub>2</sub>/nmc) fluorite structure in the ultrathin  
687 HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> films, as the presence of many reflections from the in-plane GID spectra (Fig.

688 1e, Extended Data Fig. 2a) allow for clear distinction from other nonpolar fluorite-structure  
689 polymorphs. Such diffraction spectra would be otherwise prohibited in typical out-of-plane  
690 geometry due to the lack of vertical diffraction planes and the large linewidth inherent to  
691 ultrathin films.

692  
693 **Two-dimensional diffraction** Two-dimensional reciprocal space maps (Extended Data Fig. 2b)  
694 were measured at Beamline 11-3 of the Stanford Synchrotron Radiation Lightsource, SLAC  
695 National Accelerator Laboratory. Rayonix MX225 CCD area detector collected diffraction flux  
696 in grazing incidence ( $<0.20^\circ$ ) geometry; the X-ray source (50 microns vertical x 150 microns  
697 horizontal beam size) was fixed at 12.7 keV. The sample-detector work distance was set to 80  
698 mm to enable detection of a wide region of reciprocal space ( $Q$ -range 0.2 to  $5 \text{ \AA}^{-1}$ ) at the expense  
699 of reciprocal space resolution, set by the pixel size. The two-dimensional diffraction scans – in  
700 which a wide portion of the entire reciprocal space was collected simultaneously, rather than at  
701 discrete regions in  $Q_x$ - $Q_y$  space – were averaged over data collection time and for repeated scans.  
702 These measurement features, in tandem with the high X-ray flux afforded by the synchrotron  
703 source, enabled sufficient diffraction signal detection and contrast in films just two nanometers  
704 in thickness. Data analysis was performed Nika, an Igor Pro package for correction, calibration,  
705 and reduction of two-dimensional areal maps into one-dimensional data<sup>53</sup>. Two-dimensional  
706 reciprocal space maps on bare  $\text{HfO}_2$ - $\text{ZrO}_2$ - $\text{HfO}_2$  heterostructures confirm the presence of  
707 crystalline ultrathin films despite the low deposition temperature, afforded by the low  
708 crystallization temperature of  $\text{ZrO}_2$  on  $\text{Si}$ <sup>50</sup>.

709  
710 **Ferroic phase identification from diffraction** For fluorite-structure thin films, the main phases  
711 to consider are the dielectric monoclinic ( $\text{P}2_1/\text{c}$ ), antiferroelectric tetragonal ( $\text{P}4_2/\text{nmc}$ ), and  
712 ferroelectric orthorhombic ( $\text{Pca}2_1$ ) phases. Various diffraction reflections from the wide-angle  
713 IP-GiD spectra enable indexing to the orthorhombic  $\text{Pca}2_1$  phase. Lattice parameters ( $a$ ,  $b$ ,  $c$ ) –  
714 determined via Bragg's law from the  $d_{200}$  family of reflections – are self-consistently checked  
715 against the (111) lattice spacing ( $\frac{1}{d_{111}^2} = \frac{1}{a^2} + \frac{1}{b^2} + \frac{1}{c^2}$ ) as well as other higher-order reflections  
716 present in the in-plane diffraction spectra (Extended Data Fig. 2a). For example, the lattice  
717 parameters extracted from the {200} reflections were  $a = 5.36 \text{ \AA}$ ,  $b = 5.23 \text{ \AA}$ ,  $c = 5.47 \text{ \AA}$ . This  
718 corresponds to a  $d_{211}$  lattice spacing of  $2.209 \text{ \AA}$ , which agrees well with the lattice spacing ( $2.205$   
719  $\text{ \AA}$ ) obtained from Bragg's law based on the reflection position.

720  
721 The monoclinic phase was ruled out due to a lack of two {111} peaks in the diffraction spectra  
722 and the  $(111)_o$  and  $(101)_t$  reflections being significantly offset from its expected peak position in  
723 the monoclinic phase. With regards to the indexing of tetragonal  $(101)_t$  peak (Extended Data Fig.  
724 2a), it is always reported that the tetragonal  $(101)_t$  reflection has a smaller  $d$ -spacing<sup>54</sup> in thicker  
725  $\text{HfO}_2$ -based films<sup>55</sup>, and is therefore expected to be present at a higher angle compared to the  
726 orthorhombic  $(111)_o$  reflection, which is the case in the indexed diffraction spectra (Extended  
727 Data Fig. 2a) based on the self-consistent indexing methodology outlined above provides.

728  
729 In terms of extracting the phase fraction of the tetragonal and orthorhombic phases, while  
730 Rietveld refinement has been applied to grazing incidence x-ray diffraction of thick (10 nm)  
731  $\text{Zr:HfO}_2$ <sup>56</sup> to determine the orthorhombic phase fraction, that methodology cannot be applied in  
732 the ultrathin regime, as the films are highly oriented, as opposed to fully polycrystalline  
733 (Extended Data Fig. 2b), which is a requirement to apply Rietveld refinement.

734

735 Regarding strain effects: strain-induced ferroelectricity in antiferroelectrics is a key  
736 consideration; strain-induced ferroelectricity has been predicted in  $\text{ZrO}_2$ <sup>23</sup>, which is indeed what  
737 we observe in certain lateral regions of our film. From the cross-sectional TEM (Fig. 1d,  
738 Extended Data Fig. 2e,f), the presence of both the FE orthorhombic  $\text{Pca}_21$  phase grains and AFE  
739  $\text{P4}_2/\text{nmc}$  tetragonal phase grains can be locally identified to persist throughout the entire  $\text{HfO}_2$ -  
740  $\text{ZrO}_2$ - $\text{HfO}_2$  thickness. Considering the 2 nm  $\text{HfO}_2$ - $\text{ZrO}_2$ - $\text{HfO}_2$  heterostructure has distinct layers  
741 (evidenced by XRR, EELS, and XPS characterization in Extended Data Fig. 1), that means the  
742 middle  $\text{ZrO}_2$  layer has local regions where it is stabilized in the FE orthorhombic phase, and  
743 other local regions where it is stabilized in the AFE tetragonal phase.

744

745 Regarding structural indicators of such strain effects, again we look to the measured  $d_{111}$  ( $O$ -  
746 phase) and  $d_{101}$  ( $T$ -phase) lattice spacings for the  $\sim 2$  nm  $\text{HfO}_2$ - $\text{ZrO}_2$ - $\text{HfO}_2$  (HZH) film –  
747 structural markers for distortion and strain in this fluorite-structure system<sup>13</sup>. Note that truly  
748 stress-free values cannot be obtained as bulk ferroelectricity is not stabilized in this material  
749 system, so we compare against DFT values<sup>22</sup> for  $\text{HfO}_2$ ,  $\text{ZrO}_2$ , and  $\text{Zr:HfO}_2$  (HZO)<sup>22</sup>, which  
750 closely match experimental values for thicker  $\text{HfO}_2$ - $\text{ZrO}_2$  ferroic films<sup>57</sup>.

751

752 In particular, the ferroelectric  $O$ -phase  $d_{111}$ -spacing for  $\text{HfO}_2$ - $\text{ZrO}_2$ - $\text{HfO}_2$  (3.09 Å) is larger than  
753 typical values for thick ferroelectric HZO films (2.95 Å)<sup>22</sup>, demonstrating that the individual  
754  $\text{HfO}_2$  and  $\text{ZrO}_2$  layers in the HZH multilayer are in fact strained i.e. increased rhombic distortion.  
755 This is consistent with the ultrathin-enhanced lattice distortions trend observed in previous ALD-  
756 grown highly-oriented orthorhombic ferroelectric HZO films<sup>13</sup> as well as epitaxial orthorhombic  
757 ferroelectric HZO films<sup>58</sup>. On the other hand, we observe that the  $d$ -spacing for the tetragonal  
758 (101) reflection (2.95 Å) is nearly the same as is expected for prototypical antiferroelectric  $T$ -  
759 phase  $\text{ZrO}_2$  (2.94 Å)<sup>22</sup>. This is expected: when the tetragonal phase is strained, it transitions to  
760 the lower symmetry orthorhombic phase as opposed to remaining in the tetragonal phase, as it  
761 does not have the same tolerance of the FE  $O$ -phase to maintain its symmetry when strained.  
762 Consequently, the larger  $d$ -spacing is always attributed to the FE  $O$ -phase<sup>57</sup>, as confirmed by  
763 self-consistent indexing to higher-order reflections (Extended Data Fig. 2a). These diffraction-  
764 based  $d$ -spacings are confirmed by cross-sectional TEM (Extended Data Fig. 2e,f). Furthermore,  
765 the presence of the  $\text{ZrO}_2$  layer developing ferroelectricity is supported by the presence of orbital  
766 polarization at the Zr  $L$ -edge from synchrotron X-ray linear dichroism (Extended Data Fig. 2c).

767

768 **X-ray absorption spectroscopy** Hard and soft synchrotron X-ray spectroscopy (Extended Data  
769 Fig. 2c) was measured at beamline 4-ID-D of the Advanced Photon Source, Argonne National  
770 Laboratory and Beamline 4.0.2. of the Advanced Light Source, Lawrence Berkeley National  
771 Laboratory, respectively. Spectroscopy measurements were taken at the oxygen  $K$ -edge (520-550  
772 eV), zirconium  $M_{3,2}$ -edge (325-355 eV), hafnium  $M_3$ -edge (2090-2150 eV), and zirconium  $L_{3,2}$ -  
773 edge (2200-2350 eV). X-rays were incident at 20° off grazing. XAS (XLD) was obtained from  
774 the average (difference) of horizontal and vertical linearly polarized X-rays. To eliminate  
775 systematic artifacts in the signal that drift with time, spectra measured at ALS were captured  
776 with the order of polarization rotation reversed (e.g., horizontal, vertical, vertical, and horizontal)  
777 in successive scans, in which an elliptically polarizing undulator tuned the polarization and  
778 photon energy of the synchrotron X-ray source<sup>59</sup>. Spectra measured at ALS were recorded under  
779 total electron yield (TEY) mode<sup>59</sup> from room temperature down to 100 K. Spectra measured at

780 APS were recorded under various modes: total electron yield (TEY), fluorescence yield (FY),  
781 and reflectivity (REF).

782

783 **Ferroic phase identification from spectroscopy** X-ray spectroscopy provides various  
784 signatures to distinguish the competing ferroelectric orthorhombic (Pca2<sub>1</sub>) and antiferroelectric  
785 tetragonal (P4<sub>2</sub>/nmc) phase. Simulated XAS spectra at the oxygen *K*-edge (Extended Data Fig.  
786 3d) for ZrO<sub>2</sub> in the various fluorite-structure polymorphs (orthorhombic Pca2<sub>1</sub> and tetragonal  
787 P4<sub>2</sub>/nmc) were computed through the Materials Project<sup>60</sup> open-source database for XAS  
788 spectrum<sup>61</sup>. The *T*-phase (P4<sub>2</sub>/nmc) nonpolar distortion (*D*<sub>4h</sub>, 4-fold prismatic symmetry) from  
789 regular tetrahedral (*T*<sub>d</sub>, full tetrahedral symmetry) fluorite-structure symmetry does not split the  
790 degenerate *e*-bands (*d*<sub>*x*<sup>2</sup>-*y*<sup>2</sup></sub>, *d*<sub>*3z*<sup>2</sup>-*r*<sup>2</sup></sub>), as confirmed by experiment<sup>62</sup> and the aforementioned  
791 XAS simulations<sup>13</sup>. Meanwhile, the *O*-phase (Pca2<sub>1</sub>) polar rhombic pyramidal distortion (*C*<sub>2v</sub>, 2-  
792 fold pyramidal symmetry) does split the *e*-manifold based on crystal field symmetry, providing a  
793 spectroscopic means to distinguish the *T*- and *O*-phases. The additional spectroscopic feature  
794 present between the main *e*- and *t*<sub>2</sub>- absorption features due to orthorhombic symmetry-lowering  
795 distortion is illustrated by its crystal field diagram (Extended Data Fig. 3b). This provides a  
796 spectroscopic fingerprint for phase identification beyond diffraction which can often be  
797 ambiguous due to the nearly identical *T*- and *O*-phase lattice parameters. For the 2 nm HfO<sub>2</sub>-  
798 ZrO<sub>2</sub>-HfO<sub>2</sub> trilayer, the experimental O *K*-edge XAS spectra demonstrates tetrahedral and  
799 rhombic splitting features closely matching the polar *O*-phase (Pca2<sub>1</sub>) emerge slightly below  
800 room temperature, indicative of the mixed tetragonal-orthorhombic to orthorhombic phase  
801 transition upon cooling. This temperature-dependent tetragonal-orthorhombic structural  
802 evolution is expected for fluorite-structure thin films<sup>63</sup> and is consistent with temperature-  
803 dependent capacitance measurements (Extended Data Fig. 3f). Further XAS phase identification  
804 details are provided in previous work on ultrathin Zr:HfO<sub>2</sub> films<sup>13</sup>.

805

806 **X-ray photoelectron spectroscopy** Angle-resolved photoelectron spectroscopy (ARPES) was  
807 performed using a Phi Versaprobe III at the Stanford Nano Shared Facilities (Extended Data Fig.  
808 1d). A monochromated aluminum source was used to give a photon energy of 1486.6 eV. Data  
809 was fit and analyzed using CasaXPS. Angle-dependent XPS at various incident grazing angles  
810 enabled depth-resolved composition analysis to help confirm the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> multilayer  
811 structure.

812

813 **Dielectric measurements**

814

815 **Metal-oxide-semiconductor (MOS) capacitance** Capacitance-voltage (*C-V*) measurements  
816 were performed using a commercial Semiconductor Device Analyzer (Agilent B1500) with a  
817 multi-frequency capacitance measuring unit (MFCMU). 19 micron W tips (d.c.P-HTR 154-001,  
818 FormFactor) made electrical contact within a commercial probe station (Cascade Microtech);  
819 voltage was applied to the W top electrode and the lightly-doped Si bottom electrode was  
820 grounded. To eliminate contributions from series and parasitic resistances, frequency-dependent  
821 *C-V* measurements were performed. In particular, *C-V* data was analyzed at two frequencies  
822 (100-500 kHz regime) to allow for the extraction of accurate frequency-independent *C-V* via a  
823 three-element circuit model consisting of the capacitor and the parasitic series and parallel  
824 resistors<sup>64</sup>. The frequency-independent capacitance is given by

$$C = \frac{f_1^2 C_1 (1 + D_1^2) - f_2^2 C_2 (1 + D_2^2)}{f_1^2 - f_2^2}$$

825 where  $C_i$  and  $D_i$  refer to the measured capacitance in parallel mode ( $C_p$ - $R_p$ ) and dissipation  
 826 values at frequency  $f_i$ . The dissipation factor is given by  $D = -\cot \theta$ , where  $\theta$  is the phase. In  
 827 order to maximize the accuracy of this method, it is important the dissipation factors are small  
 828 ( $\ll 1$ ) at the frequencies chosen; therefore, high frequencies were selected.

829  
 830 **Permittivity extraction** The permittivity of  $\text{Al}_2\text{O}_3$  and  $\text{HfO}_2$  dielectric layers was extracted from  
 831 thickness-dependent MOS  $C$ - $V$  measurements on lightly-doped p-substrates (Extended Data Fig.  
 832 6). In the accumulation region of the MOS  $C$ - $V$  measurements, the MOS capacitor can be  
 833 modeled as three capacitors ( $\text{Al}_2\text{O}_3$  or  $\text{HfO}_2$  dielectric layer,  $\text{SiO}_2$  interlayer, and Si space charge  
 834 layer) in series using the following equation

$$\frac{1}{C} = \frac{1}{\epsilon_0 \epsilon_{HK}} t_{HK} + \frac{1}{\epsilon_0 \epsilon_{SiO_2}} \left[ t_{SiO_2}^{phys} + \frac{t_{CL} \epsilon_{SiO_2}}{\epsilon_{Si}} \right]$$

835 where  $t_{HK}$  is the thickness of the high- $\kappa$  ( $\text{Al}_2\text{O}_3$  or  $\text{HfO}_2$ ) layer,  $t_{phys}$  is the physical  $\text{SiO}_2$   
 836 thickness, and  $t_{CL}$  is the charge layer thickness in silicon. The physical  $\text{SiO}_2$  thickness is constant  
 837 across all the thickness series ( $\text{Al}_2\text{O}_3$  and  $\text{HfO}_2$  single layers). Additionally, the capacitance  
 838 values were extracted at various values of fixed charge ( $Q = 0$  to  $-3 \mu\text{C}/\text{cm}^2$ ) which ensures that  
 839 the charge-layer thickness is constant across all thicknesses and in the accumulation region.  
 840 Therefore, the inverse capacitance at a fixed charge as a function of film thickness should result  
 841 in a line and the permittivity can be extracted from the slope. This yielded extracted  
 842 permittivities of 9 and 19 for the  $\text{Al}_2\text{O}_3$  and  $\text{HfO}_2$  thickness series, respectively, as expected for  
 843 these systems. Note that for the  $\text{HfO}_2$  thickness series, thicknesses of 6 nm and higher were used  
 844 to ensure  $\text{HfO}_2$  stabilizes in the dielectric monoclinic phase ( $\kappa \sim 18$ )<sup>22</sup>. Similarly, the permittivity  
 845 of the  $\text{HfO}_2$ - $\text{ZrO}_2$ - $\text{HfO}_2$  heterostructures was extracted from thickness-dependent MIM  $C$ - $V$   
 846 measurements (Fig. 2b). The inverse capacitance is a linear function of the film thickness, and  
 847 the permittivity can be extracted from the slope.

848  
 849 **Electrical interlayer thickness extraction** The thickness of the  $\text{SiO}_2$  interlayer was determined  
 850 not only by TEM (Extended Data Fig. 6a), but also electrically via  $C$ - $V$  measurements of both  
 851 dielectric  $\text{HfO}_2$  and  $\text{Al}_2\text{O}_3$  thickness series on  $\text{SiO}_2$ -buffered Si (Extended Data Fig. 6f). The  
 852 inverse capacitance at a fixed charge as a function of dielectric thickness should result in a line  
 853 and the capacitance-equivalent thickness (CET) of the  $\text{SiO}_2$  interlayer and Si charge layer can be  
 854 extracted from the y-intercept. By extracting the CET at different charge values, the  $Q$ - $V$  relation  
 855 of the  $\text{SiO}_2$  interlayer and Si charge layer can be calculated through the following equation

$$V - V_{fb} = \int_0^Q \frac{t_{SiO_2}^{phys} + \frac{t_{CL} \epsilon_{SiO_2}}{\epsilon_{Si}}}{\epsilon_0 \epsilon_{SiO_2}} dQ$$

856 where  $V_{fb}$  is the flatband voltage (Extended Data Fig. 6b,d). To confirm this methodology,  
 857 another method for determining the  $Q$ - $V$  relation of the  $\text{SiO}_2$  interlayer and Si charge layer was  
 858 extracted from the  $Q$ - $V$  relations of both the dielectric  $\text{HfO}_2$  and  $\text{Al}_2\text{O}_3$  thickness series. At a  
 859 fixed charge, the corresponding voltage values of each thickness were fit to a line and the y-  
 860 intercept corresponds to the voltage value for the  $\text{SiO}_2$  interlayer and Si charge layer  $Q$ - $V$  relation  
 861 (Extended Data Fig. 6c,e). As expected, both methods lead to the same extracted  $Q$ - $V$  relation  
 862 (Extended Data Fig. 6c,e), corresponding to 8 Å EOT (Extended Data Fig. 6f) – close to the  $\text{SiO}_2$   
 863 physical thickness of 8.5 Å obtained via TEM (Extended Data Fig. 6a) – based on simulated

864 TCAD  $Q$ - $V$  relations of different  $\text{SiO}_2$  thicknesses on lightly-doped Si.

865

866 **Hysteretic  $C$ - $V$  measurements** Capacitance-voltage ( $C$ - $V$ ) measurements on MIM capacitors  
867 were performed using a commercial Semiconductor Device Analyzer (Agilent B1500) with a  
868 multi-frequency capacitance measuring unit. 19 micron W tips (d.c.P-HTR 154-001,  
869 FormFactor) made electrical contact within a commercial probe station (Cascade Microtech);  
870 voltage was applied to the W top electrode and the W bottom electrode was grounded.

871

## 872 **Electrical characterization**

873

874 **Benchmarking to HKMG literature** In Fig. 3b, the leakage-equivalent oxide thickness ( $J_G$ -  
875 EOT) scaling of the negative capacitance multilayer gate stack benchmarked against reported  
876 HKMG literature includes references taken from interlayer-scavenged 2 nm  $\text{HfO}_2$ <sup>16,19,65</sup> (red),  
877 high- $\kappa$  doped  $\text{HfO}_2$ <sup>19</sup>(gray), and  $\text{SiO}_2/\text{poly-Si}^3$  (black). In Fig. 3c, the raw mobility of long-  
878 channel transistors integrating the negative capacitance multilayer gate stack (blue) is  
879 benchmarked against industry-reported long-channel transistors integrating standard 2 nm  $\text{HfO}_2$   
880 high- $\kappa$  dielectric gate stacks (red) of various EOT<sup>42</sup>. In Extended Data Fig. 8d, the normalized  
881 mobility versus EOT scaling of the negative capacitance multilayer gate stack benchmarked  
882 against reported HKMG literature includes references taken from interlayer-scavenged 2 nm  
883  $\text{HfO}_2$ <sup>16,19,42</sup> (red) and hybrid silicate-scavenged interlayer<sup>16</sup> (magenta). In the Extended Data Fig.  
884 8d inset, the  $\text{SiO}_2$  interlayer thickness versus EOT scaling scatter plot considers the 7.0 Å EOT  
885  $\text{HfO}_2$ - $\text{ZrO}_2$ - $\text{HfO}_2$  trilayer to HKMG references which employ interlayer scavenging to reduce  
886 EOT<sup>16,19,65,66</sup>.

887

888 **Transistor transfer and output characteristics** Transistor  $I_d$ - $V_g$  and  $I_d$ - $V_d$  characterization of  
889 short-channel and long-channel transistors were performed using a commercial Semiconductor  
890 Device Analyzer (Agilent B1500). 19 micron W tips (d.c.P-HTR 154-001, FormFactor) made  
891 electrical contact within a commercial probe station (Cascade Microtech); voltage was applied to  
892 the gate and drain contacts, while the source and Si substrate were grounded.

893

894 **Mobility extraction** The low-field transistor mobility for SOI transistors integrating  $\sim 2$  nm  
895  $\text{HfO}_2$ - $\text{ZrO}_2$ - $\text{HfO}_2$  ferroic multilayers and standard high- $\kappa$   $\text{HfO}_2$  gate stacks of the same physical  
896 thickness (Fig. 3c, Extended Data Fig. 8c) is calculated based on the channel resistance ( $R_{ch}$ ) and  
897 inversion sheet charge density ( $Q_{inv}$ ), which are extracted respectively from transfer  
898 characteristics ( $I_d$ - $V_{gs}$ , Extended Data Fig. 8b) and from the intrinsic gate capacitance-voltage  
899 ( $C_{gg}$  vs  $V_{gs}$ - $V_{fb}$ , Extended Data Fig. 8a) measurements. Given the device aspect ratio of channel  
900 length ( $L$ ) and channel width ( $W$ ), we have

$$R_{ch}(V_{gs}) = \frac{L}{W} \times \frac{1}{\mu_{eff}(V_{gs})Q_{inv}(V_{gs})}$$

901 Firstly, the channel resistance is extracted at 50 mV drain-to-source bias ( $V_{ds}$ ) by subtracting the  
902 parasitic resistance ( $R_p$ ) from the measured drain-to-source resistance ( $R_{ds}$ ).

$$R_{ds}(V_{gs}) = \frac{V_{ds}}{I_D(V_{gs})} = R_{ch}(V_{gs}) + R_p$$

903 where  $R_p$  is ascribed to the resistance of the source and the drain contacts and the n+ extension  
904 regions that are extrinsic to the channel region. When the overdrive voltage ( $V_{ov} = V_{gs} - V_T$ ,  
905 where  $V_T$  is the threshold voltage) is sufficiently large,  $R_{ch}$  is known to be inversely proportional



906 to  $V_{ov}$ . Therefore,  $R_p$  can be extracted using a linear extrapolation of the  $R_{ds} - 1/V_{ov}$  relationship,  
 907 which is derived from the  $I_d - V_{gs}$  (Extended Data Fig. 8b) from which  $V_T$  can be characterized  
 908 with the max- $g_m$  method. Secondly, the  $C_{gg}$  vs  $V_{gs} - V_{fb}$  (Extended Data Fig. 8a) is integrated and  
 909 normalized to the channel area to estimate the inversion charge.

$$Q_{inv}(V_{gs}) \approx \int_{-\infty}^{V_{gs}} \frac{C_{gg}(v_{gs})}{A} dv_{gs}$$

910 Finally, we combine the above characterizations to obtain the effective mobility (Fig. 3c,  
 911 Extended Data Fig. 8c).

912  
 913 **Transconductance extraction from DC measurements** The measured transconductance ( $g_m =$   
 914  $\partial I_d / \partial V_{gs}$ ) and the output conductance ( $g_{ds} = \partial I_d / \partial V_{ds}$ ) are affected by the series resistance on the  
 915 source ( $R_s$ ) and the drain sides ( $R_d$ ), as they reduce the voltage drops on the channel region,

$$\begin{aligned} V_{gs,i} &= V_{gs} - I_d R_s \\ V_{ds,i} &= V_{ds} - I_d (R_s + R_d) \end{aligned}$$

916  
 917 where  $V_{gs,i}$  and  $V_{ds,i}$  are the gate-to-source and the drain-to-source voltages intrinsic to the  
 921 channel, respectively.  $R_s \approx R_d \approx R_p/2$  because the transistor is symmetric.  $R_p$  can be extracted  
 922 from the  $R_{ds} - 1/V_{ov}$  relationships as discussed in the "Mobility Extraction" Methods section.  
 923 Besides, devices with different gate length ( $L_G$ ) series are fabricated on Silicon-On-Insulator  
 924 (SOI) wafers, which enables another extraction method with  $R_{sd} - L_G$  relations. At low  $V_d$  and a  
 925 given  $V_{ov}$ ,  $Q_{inv}$  and  $\mu_{eff}$  are unchanged across different  $L_G$  if short-channel effects are not  
 926 significant, making  $R_{ch}$  proportional to the channel length. Such condition is confirmed by the  
 927 consistency of  $V_T$  across measured  $L_G$  (Extended Data Fig. 10a). Therefore, the  $L_G$  offset as well  
 928 as the  $R_p$  can be found at the intersection of the linear relations of the  $R_{sd} - L_G$  with different  $V_{ov}$   
 929 (Extended Data Fig. 10c). The two  $R_p$  extraction methods yield consistent results.

930  
 931 The following equation is solved to extract the intrinsic  $g_{m,i} = \partial I_d / \partial V_{gs,i}$  and  $g_{ds,i} = \partial I_d / \partial V_{ds,i}$   
 932 without the degradation due to  $R_s$  and  $R_d$ .

$$\begin{pmatrix} 1 - g_m R_s & -g_m (R_s + R_d) \\ -g_{ds} R_s & 1 - g_{ds} (R_s + R_d) \end{pmatrix} \begin{pmatrix} g_{mi} \\ g_{dsi} \end{pmatrix} = \begin{pmatrix} g_m \\ g_{ds} \end{pmatrix}$$

934  
 935 where  $g_m$  and  $g_{ds}$  are measured, and  $R_s \approx R_d \approx R_p/2$  from the above discussed characterizations.  
 936 Using this methodology, the intrinsic  $g_{m,i}$  and intrinsic  $g_{ds,i}$  are extracted (Fig. 3g, Extended Data  
 937 Fig. 10d,e).

938  
 939 **Transconductance extraction from RF measurements** Scattering-parameters ( $S$ -parameters)  
 940 for  $L_G = 1 \mu\text{m}$  bulk transistors (henceforth referred to as the device under test, DUT) at various  
 941 DC biases as well as open and short structures (Extended Data Fig. 9a) are measured using a  
 942 Keysight E8361C Network Analyzer in conjunction with a Keysight 4155C Semiconductor  
 943 Parameter Analyzer. The devices were measured using low contact resistance Infinity Series  
 944 probes. To calibrate the measurement setup, a line-reflect-reflect-match (LRRM) calibration was  
 945 performed with a Cascade Microtech Impedance Standard. Following calibration,  $S$ -parameters  
 946 were measured for each of the DUT, open, and short structures. These measured  $S$ -parameters  
 947 were converted to admittance parameters ( $Y$ -parameters),  $Y_{DUT}$ ,  $Y_{open}$ , and  $Y_{short}$ . To remove the

948 effects of parasitic shunt parasitic pad capacitance and series pad resistance and inductance of the  
 949 DUT, the following de-embedding process was followed. First, to decouple the effect of shunt  
 950 parasitic capacitances, the  $Y$ -parameters of the open structure ( $Y_{open}$ ) are subtracted from the  $Y$ -  
 951 parameters of the DUT and short structure, and then are converted to impedance parameters ( $Z$ -  
 952 parameters):

$$953$$

$$954 \quad Z_1 = (Y_{DUT} - Y_{open})^{-1}$$

$$955 \quad Z_2 = (Y_{short} - Y_{open})^{-1}$$

956 Next, to decouple the effect of series pad resistance and inductance of DUT,  $Z_2$  is subtracted  
 957 from  $Z_1$  and the resulting difference is converted back to admittance parameters,  $Y_{corr}$ :

$$958$$

$$959 \quad Y_{corr} = (Z_1 - Z_2)^{-1}$$

960

961  $Y_{corr}$  represents the de-embedded admittance parameters of the DUT. This de-embedding  
 962 procedure is schematically represented in Extended Data Fig. 9a.

963

964 To extract the transconductance ( $g_m$ ) from the de-embedded admittance parameters, a small-  
 965 signal model of the transistor was assumed (Extended Data Fig. 9b). Under this small-signal  
 966 model, the  $Y$ -parameters can be written in terms of model parameters and frequency (assuming  
 967  $R_s = R_d = 0$ ,  $C_{gg} = C_{gs} + C_{gd}$ , and  $4\pi^2 C_{gg}^2 R_g^2 f^2 \ll 1$ )

$$968$$

$$969 \quad Y_{11} = 4\pi^2 C_{gg}^2 R_g f^2 + j2\pi f C_{gg}$$

$$970 \quad Y_{12} = -4\pi^2 C_{gd} C_{gg} R_g f^2 - j2\pi f C_{gd}$$

$$971 \quad Y_{21} = g_m - 4\pi^2 C_{gd} C_{gg} R_g f^2 + j2\pi f (C_{gd} + g_m R_g C_{gg})$$

$$972 \quad Y_{22} = g_{ds} + 4\pi^2 C_{gd} R_g (C_{gd} + C_{gg} g_m R_g) f^2 + j2\pi f (C_{ds} + C_{gd} + C_{gd} g_m R_g).$$

973

974 The transconductance ( $g_m$ ) can therefore be extracted at a fixed DC bias via the following  
 975 relation (Fig. 3h, Extended Data Fig. 9c).

$$976 \quad g_m = \text{Re}(Y_{21})|_{f^2=0}$$

977 **Reliability** Positive bias temperature instability (PBTI) measurements were performed on bulk  
 978 nMOSFET devices integrating the  $\sim 2$  nm mixed-ferroic  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  (HZH) and  
 979 conventional high- $\kappa$  dielectric  $\text{HfO}_2$  gate stacks at  $85^\circ\text{C}$  at electric fields up to 9 MV/cm  
 980 (Extended Data Fig. 8f,g). A measure-stress-measure (MSM) voltage scheme (Extended Data  
 981 Fig. 8e) was used to apply the PBTI bias, where the drain current was measured with a  
 982 minimized delay time (600  $\mu\text{s}$ ) at  $V_{ds} = 50$  mV to minimize the recovery effect<sup>67</sup>. The measured  
 983 drain current was then converted to a  $\Delta V_T$  shift by comparing it to the drain current measured on  
 984 the virgin device. Additionally, the time exponent,  $n$ , was extracted by noting that<sup>67</sup>  $\Delta V_T = At^n$ .  
 985 The extracted time exponent,  $n$ , was found to similar to those reported in literature for high- $\kappa$   
 986  $\text{HfO}_2$  stacks<sup>67</sup>, which is expected considering the reliability characteristics are predominantly  
 987 determined by the interfacial (IL) oxide and IL-high- $\kappa$  interface<sup>46</sup>; both stacks with different  
 988 EOT have  $\text{HfO}_2$  sitting on the same  $\text{SiO}_2$  IL (Extended Data Fig. 1). Furthermore, the DC  
 989 lifetime<sup>67</sup> – the stress time needed to induce a 50 mV  $\Delta V_T$  shift – was extracted as a function of  
 990 electric field from the PBTI measurements for the HZH and  $\text{HfO}_2$  gate stacks. Both HZH and  
 991  $\text{HfO}_2$  show comparable rates of degradation as a function of field (Extended Data Fig. 8h), which  
 992 is expected for the aforementioned reasons related to the consistent  $\text{SiO}_2$  IL.

993

994 Additionally, the extracted time exponent for HZH ( $n = 0.14$ , Extended Data Fig. 8f) is closer to  
995 the ideal value<sup>67</sup> of  $n = 0.16$  compared to HfO<sub>2</sub> ( $n = 0.10$ , Extended Data Fig. 8g), indicating that  
996 there are initially a smaller number of interface traps for HZH. When field stress is applied, trap  
997 generation accelerates until the number of traps reaches a certain threshold beyond which it  
998 eventually saturates. As a result, the relative degradation is larger for HZH at smaller fields,  
999 although the absolute degradation is always slightly smaller than HfO<sub>2</sub>. This can also be seen  
1000 directly from the extracted DC lifetimes (Extended Data Fig. 8h) as the DC lifetime is slightly  
1001 better for HZH at intermediate field stresses before it becomes similar to HfO<sub>2</sub> at high field  
1002 stresses. We again note that extracted  $n$  values are similar to what has been reported in literature  
1003 for HfO<sub>2</sub> based high- $\kappa$  metal gate stacks<sup>67</sup>.

1004

1005 Stress measurements were also performed on lightly-doped p-type MOS capacitors with the  $\sim 2$   
1006 nm mixed-ferroic HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> and conventional high- $\kappa$  dielectric HfO<sub>2</sub> gate stacks at room  
1007 temperature (Extended Data Fig. 8i,j) at  $V_g - V_{fb} = -1$  V. The stresses were applied again with a  
1008 MSM voltage scheme, where the accumulation  $C$ - $V$  was measured in between bias application at  
1009 500 kHz. The stress-induced effect was found to be minimal (Extended Data Fig. 8i,j) and no  
1010 EOT degradation was observed after  $10^3$  s of stress at  $V_g - V_{fb} = -1$  V (Extended Data Fig. 8i,j).

1011

1012 **Charge boost measurements** Pulsed charge-voltage measurements (Extended Data Fig. 7)  
1013 were conducted on p-Si/SiO<sub>2</sub>/HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> (2 nm)/TiN/W capacitor structures to extract the  
1014 energy landscape of the ferroic HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructure, following the measurement  
1015 scheme detailed in previous works<sup>26,68-70</sup>. The capacitor structures were connected to an Agilent  
1016 81150A Pulse Function Arbitrary Noise Generator and the current and voltage was measured  
1017 through an InfiniiVision DSOX3024A oscilloscope with 50  $\Omega$  and 1 M $\Omega$  input impedances,  
1018 respectively. Short voltage pulses (500 ns) with increasing amplitudes were applied to the  
1019 capacitor (Extended Data Fig. 7c). From the integration of the measured discharging current, a  
1020 charge vs voltage relationship was extracted (Extended Data Fig. 7d). The voltage was calculated  
1021 by  $\max(V - IR)$ , where  $V$  is the applied voltage pulse,  $I$  is the measured current, and  $R$  is a  
1022 combination of the oscilloscope resistance (50  $\Omega$ ) and parasitic resistances associated with the  
1023 setup and lightly-doped substrate (220  $\Omega$ ). Fast voltage pulses were applied in order to minimize  
1024 charge injection into the ferroelectric-dielectric interface, which could mask the observation of  
1025 the negative capacitance regime<sup>26,69</sup>. Additionally, short voltage pulses help prevent electrical  
1026 breakdown of the SiO<sub>2</sub> layer. The  $Q$ - $V$  relation of the series capacitance of the SiO<sub>2</sub> interlayer  
1027 and Si charge layer was determined via thickness-dependent  $C$ - $V$  measurements of Al<sub>2</sub>O<sub>3</sub> and  
1028 HfO<sub>2</sub> (Extended Data Fig. 6, Methods, Electrical interlayer thickness extraction), which  
1029 corresponded to 8 Å SiO<sub>2</sub> on lightly-doped Si. The charge boost was calculated by integrating  
1030 the difference between the  $Q$ - $V$  relations of the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructure and the  
1031 series combination of the SiO<sub>2</sub> interlayer and the Si charge layer (Extended Data Fig. 7e).

1032

1033 In order to determine the polarization-electric field ( $P$ - $E_F$ ) relation of just the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-  
1034 HfO<sub>2</sub> heterostructure (Extended Data Fig. 7f), the electric field across the ferroic HfO<sub>2</sub>-ZrO<sub>2</sub>-  
1035 HfO<sub>2</sub> heterostructure was calculated by subtracting the voltage across the series capacitance of  
1036 the SiO<sub>2</sub> interlayer and Si charge layer ( $V_D$ ) at a fixed charge value,

$$E_F = \frac{1}{t}(V - V_D)$$

1037 where  $t$  is the thickness of the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructure.

1038

## 1039 **Modeling**

1040

1041 **Energy landscape considerations** One can write the total free energy ( $F$ ) of the system as:

1042

$$F = \int_V [f_{bulk} + f_{elas} + f_{elec} + f_{grad}] dV$$

1043 where  $V$  is the volume,  $f_{bulk}$  is the bulk free energy (Landau),  $f_{elas}$  is the elastic energy density,  $f_{elec}$   
1044 is the electrostatic energy density, and  $f_{grad}$  is the gradient energy density.

1045

1046 For the laterally-arranged mixed FE-AFE phase present in our material, all of the above terms  
1047 are important, especially the gradient terms, which are by default present due to the mixed polar-  
1048 nonpolar (FE-AFE) phase distribution. Additionally, heterogeneous elastic energies in  
1049 structurally inhomogeneous systems – like our mixed orthorhombic-tetragonal (FE-AFE) system  
1050 – have been shown to destabilize long-range polarization, leading to suppressed polarization and  
1051 a flattened energy landscape<sup>28,71</sup>. Furthermore, considering the polarization in our films has an  
1052 in-plane component (as described in the Main Text), this leads to an additional depolarization  
1053 field on the ferroelectric grains, similar to a ferroelectric-dielectric heterostructure (albeit in the  
1054 in-plane direction). At low electric fields, the mixed ferroelectric-antiferroelectric behavior is  
1055 analogous to a ferroelectric-dielectric (polar-nonpolar) heterostructure – due to the nonpolar  
1056 parent structure of fluorite-structure antiferroelectricity – which has been shown to impart  
1057 depolarization fields on the ferroelectric layer<sup>25,72</sup>. The laterally-intertwined nonpolar-polar  
1058 phases present in the ultrathin HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> heterostructure are conducive to flattening the  
1059 ferroelectric energy landscape through the aforementioned depolarization fields<sup>26–28</sup> (Fig. 1a).

1060

1061 Overall, the above contributions all lead to a suppression of the bulk polarization via  
1062 depolarization fields. As it has been shown<sup>26–28</sup>, the depolarization field essentially flattens the  
1063 bulk energy landscape for the ferroelectric – ( $E_d \propto -P$  hence  $\vec{E} \cdot \vec{P} > 0$ ) – and leads to a  
1064 permittivity enhancement ( $\epsilon \propto [\partial^2 F / \partial D^2]^{-1}$ ). Depolarization field induced flattening of the energy  
1065 landscape is also the underlying physics of the negative capacitance effect<sup>26,27,29,30,49</sup>.

1066

1067 **Technology computer-aided design simulations** The measured  $C$ - $V$  curves are calibrated to  
1068 Sentaurus Technology computer-aided design simulations (TCAD) device simulator which  
1069 solves the electrostatics, electron and hole transports, and the quantum confinement effect self-  
1070 consistently<sup>73</sup>. MOS capacitors with  $10^{15}$  cm<sup>-3</sup> p-type substrate doping and planar SOI  
1071 MOSFETs are simulated with finite-element methods. The equivalent oxide thickness (EOT) and  
1072 the metal work function ( $\phi_m$ ) are the only two parameters that are fit to the MOS capacitor  
1073 measurement results, yet the slope of the accumulation capacitance can be successfully captured  
1074 by the model (Fig. 2f, Extended Data Fig. 6). Similarly, the intrinsic  $C_{gg}$  vs  $V_{gs} - V_{fb}$  extracted  
1075 from SOI transistors can be successfully model by the TCAD model with appropriate EOT and  
1076  $\phi_m$  (Extended Data Fig. 8a).

1077

## 1078 **Atomic-scale HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> mixed-ferroic heterostructure**

1079

1080 **Thickness limits and atomic-scale heterostructures** Recent perspectives on HfO<sub>2</sub>-based

1081 ferroelectricity for device applications<sup>9,74-77</sup> posed the technological challenges stemming from  
1082 thickness limit concerns of HfO<sub>2</sub>-based ferroelectricity, and thereby, negative capacitance. The  
1083 use of short-period superlattices i.e. nanolaminates is common in the high- $\kappa$  field to enhance per-  
1084 mittivity<sup>78-82</sup>; in particular, rutile-structure TiO<sub>2</sub> is often paired with fluorite-structure HfO<sub>2</sub>  
1085 and/or ZrO<sub>2</sub> in DRAM capacitors<sup>83</sup>. Recently, fluorite-structure nanolaminates were employed to  
1086 tune the ferroelectric behavior of HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> films<sup>84-86</sup>. However, all of these works have  
1087 studied nanolaminates with thick periodicity, going as thin as 10 ALD cycles ( $\sim$  1.1 nm) per  
1088 superlattice sub-layer<sup>84</sup>. In this work, we scale down to a much thinner thickness limit while still  
1089 maintaining physical separation of the individual layers (Extended Data Fig. 1). The reasoning  
1090 behind using a short-period superlattice structure to scale down the ferroic behavior of HfO<sub>2</sub>-  
1091 ZrO<sub>2</sub>-HfO<sub>2</sub> rather than simply thinning down a solid solution stems from the notorious thickness-  
1092 dependent ferroelectric behavior in Zr:HfO<sub>2</sub> at fixed composition<sup>38,57,63</sup>. Here, the use of  
1093 nanolaminated structures can help provide thickness-independent scaling of ferroic order, as has  
1094 been previously demonstrated to overcome the upper thickness limit of HfO<sub>2</sub>-based  
1095 ferroelectricity<sup>86</sup>. The persistence of high capacitance for these 2 nm films is notable considering  
1096 other high- $\kappa$  dielectric systems suffer from significant permittivity degradation in the thin film  
1097 (sub-10 nm) regime, particularly TiO<sub>2</sub>- and SrTiO<sub>3</sub>-based oxides<sup>83,87</sup>. Sustaining the mixed  
1098 ferroic order underlying negative capacitance to the 2 nm regime is extremely relevant for  
1099 advanced technology nodes<sup>88</sup> which budget only  $\sim$  2 nm for the oxide layer.

1100

1101 **Iso-structural polycrystalline multilayer** Previous attempts to heterostructure ferroelectric  
1102 Zr:HfO<sub>2</sub> with dielectric Al<sub>2</sub>O<sub>3</sub><sup>26,69,70</sup> failed to demonstrate capacitance enhancement, which was  
1103 attributed to the fixed charges at the ferroelectric-dielectric interface. These charges can screen  
1104 the ferroelectric polarization, pushing the stable point of the energy well to one the minimum  
1105 points, and thereby preventing stabilization of negative capacitance regime via depolarization  
1106 fields from the dielectric. Here, the use of iso-structural HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> to serve as both the  
1107 nonpolar (antiferroelectric) and polar (ferroelectric) layers and leveraging the high (low) onset  
1108 crystallization temperature of HfO<sub>2</sub> (ZrO<sub>2</sub>) on Si<sup>50</sup>, enables interfaces with diminished defects,  
1109 allowing for the polar layer to experience the depolarization fields and stabilize in the  
1110 "forbidden" NC regime. Regarding the polycrystalline nature of the ultrathin multilayers, it has  
1111 been experimentally<sup>48</sup> and theoretically<sup>89</sup> established that negative capacitance can be stabilized  
1112 in the presence of ferroelectric domains, as recently reviewed<sup>76</sup>.

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1223 **Data availability** The experimental data contained in the manuscript are available for download  
1224 at <https://doi.org/10.5281/zenodo.5797030>.  
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1226 **Extended Data Fig. 1. Atomic-scale multilayer structure.** (a) Schematic of the HfO<sub>2</sub>-ZrO<sub>2</sub>  
1227 multilayer structure on SiO<sub>2</sub>-buffered Si. (b) Synchrotron x-ray reflectivity (XRR) of thicker  
1228 HfO<sub>2</sub>-ZrO<sub>2</sub> heterostructures (left) repeated with the same periodicity as the thinner trilayer  
1229 structure; XRR fitting (right) demonstrates the presence of well-separated HfO<sub>2</sub>-ZrO<sub>2</sub> layers, i.e.  
1230 not a solid- solution, for three different multilayer repeats of fixed periodicity, all approximately  
1231 following the expected 4 Å - 12 Å - 4 Å HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> structure. (c) Layer-resolved electron  
1232 energy loss spectroscopy (EELS) of the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> trilayer, demonstrating clear  
1233 separation of HfO<sub>2</sub> and ZrO<sub>2</sub> layers. The exact layer thicknesses are extracted from XRR, which  
1234 spans a wider sample footprint, rather than the local EELS measurement in which the apparent  
1235 width increase can be due to beam spreading and local thickness variation. (d) Angle-resolved  
1236 X-ray photoelectric spectroscopy (XPS) of the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> trilayer (left) and the  
1237 extracted atomic composition (right). The presence of increasing Zr-content as the grazing angle  
1238 increases is expected from the multilayer structure in which Zr-content increases after the surface  
1239 Hf-rich layer. Therefore XRR, EELS, and XPS data all indicate the presence of a multilayer  
1240 structure in which the HfO<sub>2</sub> layer is directly on the SiO<sub>2</sub> layer.

1241 **Extended Data Fig. 2. Ferroic phase insights from structural characterization.** (a) (left) In-  
1242 plane synchrotron grazing-incidence diffraction (IP-GiD) of a bare 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub>  
1243 trilayer indexed to the tetragonal P4<sub>2</sub>/nmc and orthorhombic Pca2<sub>1</sub> phases and (right) zoom-in of  
1244 the spectrum about the orthorhombic (111)<sub>o</sub> and tetragonal (101)<sub>t</sub> reflections, confirming the co-  
1245 existing structural polymorphs in the 2 nm film. These two peaks were differentiated via self-  
1246 consistent indexing of the entire spectrum, in which interplanar lattice spacings – determined  
1247 from the {200}<sub>o</sub> family of reflections – closely match the *d*-spacings for all other reflections –  
1248 (111)<sub>o</sub>, (120)<sub>o</sub>, (211)<sub>o</sub>, (202)<sub>o</sub> – determined by Bragg’s law (Methods). (b) Two-dimensional  
1249 reciprocal space map of the bare 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> trilayer, indexed by integrating the  
1250 diffraction spectrum. The lack of fully polycrystalline rings illustrates that the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-  
1251 HfO<sub>2</sub> trilayer is highly-oriented, consistent with TEM imaging. (c) Synchrotron spectroscopy  
1252 (XAS) of the bare 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> trilayer at the (left) Hf *M*<sub>3</sub>- and (center) Zr *L*<sub>3,2</sub>-edges:  
1253 (right) the presence of linear dichroism (orbital polarization) provides further evidence of  
1254 symmetry-breaking in these oriented thin films. (d) Second harmonic generation (SHG) mapped  
1255 across the bare 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> trilayer; the presence of SHG intensity confirms broken  
1256 inversion symmetry in these ultrathin ferroic films. (e, f) Additional cross-sectional TEM  
1257 providing complementary evidence of the (e) tetragonal P4<sub>2</sub>/nmc and (f) orthorhombic Pca2<sub>1</sub>  
1258 phases, in which the extracted (101)<sub>t</sub> lattice spacing (~ 2.95 Å) and (111)<sub>o</sub> lattice spacing (~ 3.08  
1259 Å) extracted from IP-GiD are consistent with the average lattice spacings extracted from the  
1260 periodicity of the TEM-imaged planes. The white scale bars in all the TEM images represent 1  
1261 nm.

1262 **Extended Data Fig. 3. Ferroic phase insights: proximity to temperature-dependent phase**  
1263 **transition.** (a) Schematic of temperature-dependent antiferroelectric-ferroelectric phase  
1264 evolution in fluorite-structure oxides. At lower temperatures, the higher symmetry tetragonal  
1265 phase is expected to transition to the lower symmetry orthorhombic phase. (b) Schematic crystal  
1266 field splitting diagram for fluorite-structure polymorphs; symmetry-induced *e*-splitting provides  
1267 a spectroscopic signature for the polar *O*-phase (Methods). (c) Temperature-dependent XAS at  
1268 the oxygen *K*-edge for a 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> bare film demonstrating clearer spectroscopic  
1269 signatures of the ferroelectric *O*-phase emerge slightly below room temperature. (d) Simulated  
1270 oxygen *K*-edge XAS spectra (Materials Project) for the respective *O*- and *T*-phases. XAS  
1271 provides spectroscopic signatures to distinguish between the *O*- and *T*-phases (difficult to resolve  
1272 from GI-XRD). (e) Prototypical *C-V* behavior for mixed antiferroelectric-ferroelectric (shoulder-  
1273 like features in addition to the characteristic butterfly-like shape) and ferroelectric films (just  
1274 butterfly-like) in MIM capacitor structures. (f) Temperature-dependent *C-V* for thicker HfO<sub>2</sub>-  
1275 ZrO<sub>2</sub> multilayers of the same periodicity (in MIM capacitor structure) demonstrating an  
1276 evolution from mixed-ferroic to ferroelectric-like hysteresis upon cooling slightly below room  
1277 temperature. Thinner HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers films suffer from leakage-limitations, preventing  
1278 such hysteretic *C-V* measurements. The thicker HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers of the same periodicity –  
1279 annealed at the same low-temperature condition to maintain the multilayer structure –  
1280 demonstrate a similar mixed ferroic to ferroelectric phase transition slightly below room  
1281 temperature as the thinner 2 nm multilayer (c).

1282 **Extended Data Fig. 4. Solid solutions versus superlattice structure: Role of ALD period and**  
1283 **Zr-content. (a)** Schematic of HfO<sub>2</sub>-ZrO<sub>2</sub> multilayer and Zr-rich Hf:ZrO<sub>2</sub> solid solution films.  
1284 With shorter ALD periods, the mixed FE-AFE multilayer structure transitions towards a Hf:ZrO<sub>2</sub>  
1285 solid-solution with AFE-like behavior. In the solid-solution state, the loss of the mixed ferroic  
1286 order yields diminished capacitance due to the lack of mixed-ferroic-induced capacitance  
1287 enhancement (Fig. 1a). **(b)** MOS accumulation *C-V* of the HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> trilayer (60% Zr)  
1288 compared to solid solutions films of the same thickness (2 nm) and composition (60% Zr), as  
1289 well as solid solutions films of the same thickness and higher Zr-composition (67%-100% Zr).  
1290 **(c)** MIM *C-V* hysteresis loops of the HfO<sub>2</sub>-ZrO<sub>2</sub> superlattice (60% Zr) compared to solid  
1291 solutions films of the same thickness (6 nm) and composition (60% Zr), as well as solid solutions  
1292 films of the same thickness and higher Zr-composition (67%-100% Zr). Hf:ZrO<sub>2</sub> solid solution  
1293 films with higher Zr-content (60%-75%) is around the range attributed to the "MPB" in thicker  
1294 Hf:ZrO<sub>2</sub> alloys<sup>35,55,90-93</sup>. These results indicate the capacitance enhancement in multilayer films is  
1295 not simply driven by Zr-content<sup>32,38,57,63</sup>, but instead the atomic-scale stacking, as the solid  
1296 solution films with sub-atomic superlattice period do not demonstrate the same mixed ferroic  
1297 behavior and enhanced capacitance as the superlattices.

1298 **Extended Data Fig. 5. Solid solutions versus superlattice structure: Role of annealing**  
1299 **temperature.** (a) Schematic of HfO<sub>2</sub>-ZrO<sub>2</sub> multilayer and Hf:ZrO<sub>2</sub> solid solution films. Under a  
1300 high-temperature anneal, the multilayer structure transitions towards a Hf:ZrO<sub>2</sub> solid-solution-  
1301 like structure demonstrating more FE-like behavior. The solid solution state yields diminished  
1302 capacitance due to the lack of both the higher-permittivity AFE phase and the mixed-ferroic-  
1303 induced capacitance enhancement (Fig. 1a). (b) Comparison of MOS capacitor accumulation C-  
1304 V characteristics in HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers, where the superstructure was repeated (left) 1,  
1305 (center) 2, or (right) 3 times, under both low- and high-temperature anneals. (c) Comparison of  
1306 mixed-ferroic behavior in low-temperature treated MIM HfO<sub>2</sub>-ZrO<sub>2</sub> multilayers versus FE  
1307 behavior in the same multilayers annealed at high temperatures, where the superstructure was  
1308 repeated (left) 3, (center) 4, or (right) 5 times. In all instances, the high-temperature anneal (>  
1309 500°C) results in diminished accumulation capacitance compared to the low-temperature  
1310 anneals, as the multilayered mixed-ferroic films presumably transition to more FE-like solid-  
1311 solution alloys.

1312 **Extended Data Fig. 6. SiO<sub>2</sub> interlayer thickness.** (a) Wide field-of-view (FoV) cross-sectional  
1313 TEM images of the HfO<sub>2</sub>-ZrO<sub>2</sub> multilayer structure and its corresponding intensity line scan  
1314 (bottom right) averaged across the entire top cross-sectional image FoV (~ 150 nm, teal-colored  
1315 box). Note the vertical teal-colored lines in the intensity line scan correspond to the inner teal-  
1316 colored box in the wide-FoV image, which delineate the SiO<sub>2</sub> interlayer boundaries. The bottom  
1317 cross-sectional TEM image highlights the thin SiO<sub>2</sub> interlayer (white region) without  
1318 obfuscation by the teal-colored box. A physical SiO<sub>2</sub> thickness of 8.6 Å is extracted from  
1319 analysis of the averaged intensity line scan of the wide FoV TEM (Methods). (b), (d) *C-V*  
1320 measurements of HfO<sub>2</sub> (b) and Al<sub>2</sub>O<sub>3</sub> (d) thickness series in MOS capacitor structures (left),  
1321 extracted inverse capacitance versus thickness at different charge values (center), and extracted  
1322 *Q-V* relation Si charge layer and SiO<sub>2</sub> interlayer (SiL) (right), which fits to TCAD simulations  
1323 for 8.0 Å SiO<sub>2</sub>. The SiL *Q-V* relation was found by integrating the extracted capacitance  
1324 equivalent thickness of SiL versus charge (right, inset). This electrical interlayer thickness (8.0  
1325 Å) is slightly less than the physical thickness determined by TEM (8.6 Å). As a sanity check, the  
1326 extracted permittivity from this methodology for HfO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> corresponds to 19 and 9,  
1327 respectively, as is expected (Methods, Permittivity Extraction). (c), (e) *Q-V* curves for HfO<sub>2</sub> (c)  
1328 and Al<sub>2</sub>O<sub>3</sub> (e) thickness series obtained from integrating MOS *C-V* measurements (left),  
1329 extracted voltage vs thickness at various charge values (center), and extracted *Q-V* relation of  
1330 SiL (right). The SiL *Q-V* relation is consistent with the *Q-V* relation extracted from the *C-V* data  
1331 (inset). (f) Consistency in the SiL *Q-V* relation extracted from the *C-V* data from both the HfO<sub>2</sub>  
1332 and Al<sub>2</sub>O<sub>3</sub> thickness series, which both fit to 8.0 Å SiO<sub>2</sub> interlayer thickness.

1333 **Extended Data Fig. 7. Capacitance and charge enhancement.** (a) MOS schematic of the 20 Å  
1334 HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> (HZH) mixed ferroic trilayer sample on lightly-doped Si (10<sup>15</sup> cm<sup>-3</sup>)  
1335 considered for the following *C-V* and pulsed *I-V* measurements. (b) Accumulation *C-V* curves  
1336 for 2 nm HZH grown on sub-nm SiO<sub>2</sub> fit to equivalent oxide thickness (EOT) simulations  
1337 (Methods). Inset: Externally verified MOS accumulation *C-V* of the same trilayer stack  
1338 (Methods), demonstrating 6.5 Å EOT. The 2 nm trilayer on top of SiO<sub>2</sub> demonstrates lower EOT  
1339 than the thickness of SiO<sub>2</sub> interlayer alone, carefully extracted via physical (8.5 Å) and electrical  
1340 (8.0 Å) methodologies (Extended Data Fig. 6), providing evidence of capacitance enhancement.  
1341 (c) The applied voltage pulse (top), measured current response (center), and integrated charge  
1342 (bottom) as a function of time for 2 nm HZH in MOS capacitors. (d) The maximum charge  $Q_{max}$ ,  
1343 the residual charge  $Q_{res}$ , and their difference,  $Q_{rev}$ , derived from the charge vs time curve for each  
1344 of the voltage pulses (Methods). (e) The reversible charge of the MOS layer (top) compared  
1345 against the extracted charge of the Si charge layer plus SiO<sub>2</sub> interlayer (SiL) derived electrically  
1346 (Extended Data Fig. 6f). The charge boost (bottom) present in the total MOS structure (SiL plus  
1347 HZH capacitors) compared to just the SiL is a signature of negative capacitance (NC)<sup>26,69</sup>. (f)  
1348 The polarization-electric field ( $P-E_F$ ) relationship for just the 2 nm HZH layer, extracted from  
1349 the charge-voltage relationship in (e). Note the presence of a negative slope regime in the  
1350 extracted  $P-E_F$  relation corresponds to NC stabilization<sup>26,69</sup>. (g) Scatter plot of reported  
1351 ferroelectric-dielectric systems demonstrating capacitance or charge enhancement at the  
1352 capacitor-level, via *C-V* or pulsed *I-V* measurements, respectively. The plot considers fluorite-  
1353 structure bilayers<sup>26,69</sup> (red), perovskite-structure bilayers<sup>29,94</sup> (blue, BL), and perovskite-structure  
1354 superlattices<sup>30,31,47,48</sup> (blue, SL). This work marks the thinnest demonstration of NC.



1355 **Extended Data Fig. 8. Mobility and Reliability** (a) Intrinsic  $C_{gg}$  vs  $V_{gs}-V_{fb}$  for  $\sim 20$  Å HfO<sub>2</sub>-  
1356 ZrO<sub>2</sub>-HfO<sub>2</sub> (HZH) and HfO<sub>2</sub> gate stacks, which fit to 7.5 Å and 9.5 Å, respectively, extracted  
1357 from SOI transistors. (b)  $I_d$  vs  $V_{gs} - V_T$  at  $V_d= 50$  mV for transistors implementing HZH and  
1358 HfO<sub>2</sub> gate stacks. (c) EOT and mobility (at  $5 \times 10^{12}/\text{cm}^2$ ) for HZH and HfO<sub>2</sub>, demonstrating no  
1359 mobility degradation. (d) Normalized mobility versus EOT for transistors integrating the 2 nm  
1360 HZH mixed-ferroic gate stack (blue) versus a 2 nm HfO<sub>2</sub> standard high- $\kappa$  dielectric gate stack  
1361 (black) of higher EOT, demonstrating no mobility degradation. These results are also  
1362 benchmarked against reported HKMG literature<sup>3</sup> implementing interlayer-scavenged 2 nm HfO<sub>2</sub>  
1363 (red). Inset: SiO<sub>2</sub> interlayer thickness versus EOT for 6.5 Å EOT HZH stack against notable  
1364 HKMG literature employ interlayer scavenging<sup>3</sup>. This scatter plot highlights the underlying  
1365 reason for the improved leakage-EOT and mobility-EOT behavior in the ultrathin HZH gate  
1366 stacks: achieving low EOT without reducing the SiO<sub>2</sub> interlayer thickness. (e) Measure-Stress-  
1367 Measure scheme used in PBTI reliability measurements. (f, g)  $\Delta V_T$  vs stress time for long-  
1368 channel bulk transistors integrating HZH (f) and HfO<sub>2</sub> (g) at varying stress conditions (up to 9  
1369 MV/cm), measured at 85°C. The extracted time exponent,  $n$ , is similar to reported high- $\kappa$  HfO<sub>2</sub>  
1370 stacks<sup>67</sup>, which is expected considering the similar interfacial (IL) oxide and IL-high- $\kappa$   
1371 interface<sup>46</sup>; both stacks with different EOT have HfO<sub>2</sub> sitting on the same SiO<sub>2</sub> IL (Extended  
1372 Data Fig. 1). (h) DC lifetime (stress time needed to induce a 50 mV  $\Delta V_T$  shift) vs electric field  
1373 for HZH and HfO<sub>2</sub>, demonstrating similar rates of degradation. (i, j) Evolution of MOSCAP  
1374 accumulation  $C-V$  curves as a function of stress time at a stress of  $V_{fb} - 1$  V for HZH (i) and HfO<sub>2</sub>  
1375 (j).  
1376  
1377

1378 **Extended Data Fig. 9. RF device characterization.** (a) De-embedding procedure for extracting  
1379 corrected admittance parameters ( $Y_{corr}$ ) by decoupling parasitic shunt capacitance and series  
1380 resistance and inductance by measuring scattering parameters for the device under test (DUT) as  
1381 well as open and short structures. More details can be found in the Methods. (b) Small-signal  
1382 model for transistor used to extract transconductance ( $g_m$ ) and total gate capacitance ( $C_{gg} = C_{gs} +$   
1383  $C_{gd}$ ). (c) De-embedded  $(2\pi f)^{-1}Re(Y_{21})$  points extrapolated to the zero frequency limit (dotted  
1384 lines) to extract the RF  $g_m$ . All data shown were extracted from bulk transistors ( $L_G = 1 \mu\text{m}$ )  
1385 integrating the 2 nm HfO<sub>2</sub>-ZrO<sub>2</sub>-HfO<sub>2</sub> ferroic gate stack.  
1386

1387 **Extended Data Fig. 10. Transconductance extraction.** (a) Threshold voltage extraction by  
1388 linear extrapolation for various channel lengths. All channel lengths give nearly constant  $V_T$  ( $\sim$   
1389 0.42 V), satisfying the assumption for the line resistance method. (b) Source/drain series  
1390 resistance extracted using the  $I/V_{ov}$  method (Methods). By performing a linear interpolation of  
1391 the total resistance for  $V_{ov} = 0.5-0.6$  V, the extracted series resistance is  $\sim 500 \Omega\text{-}\mu\text{m}$ . (c)  
1392 Source/drain series resistance extracted using the line resistance method (Methods). The trend is  
1393 considered down to  $L_G = 90$  nm, which intersects at  $\sim 500-600 \Omega\text{-}\mu\text{m}$  – consistent with the  $I/V_{ov}$   
1394 method– with an  $L_G$  offset of  $\sim 50$  nm. (d, e) Measured (left) and extracted (right)  
1395 transconductance (d) and output conductance (e) versus  $V_g$  for  $V_{ds} = 0.9-1.1$  V, assuming  $R_s = R_d$   
1396 =  $250 \Omega\text{-}\mu\text{m}$  for  $L_G = 90$  nm. The de-embedding of intrinsic  $g_{m,i}$  and  $g_{ds,i}$  from extrinsic  $g_m$  and  
1397  $g_{ds}$  is described in the Methods. All data shown were measured on SOI short-channel transistors  
1398 integrating the 2 nm  $\text{HfO}_2\text{-ZrO}_2\text{-HfO}_2$  ferroic gate stack.





