Hf$_{1-x}$Zr$_x$O$_2$ ($x = 0, 0.31, 0.46, 0.79,$ and 1) films were grown to a thickness of ~20 nm using Plasma-Enhanced Atomic Layer Deposition (PEALD) in an Oxford FlexAL II system. Tetrakis(ethylmethylamido)hafnium (TEMA-Hf) and tetrakis(ethylmethylamido)zirconium (TEMA-Zr) precursors, held at temperatures of 85 °C and 70 °C, respectively, were deposited in 10 cycle super-cycles, varying the ratio of TEMA-Hf:TEMA-Zr pulses to control composition. For TEMA-Hf pulses, 250 sccm of Ar was used as a carrier/bubbler gas, whereas 350 sccm of Ar was utilized for TEMA-Zr pulses. For both pulses, the chamber pressure was kept at 80 mTorr, and throughout the deposition the sample temperature was kept at 260 °C. For both precursors, 90 seconds of purge time was allotted after each pulse step, during which 200 sccm of Ar was flowed through the chamber at a pressure of 15 mTorr. Following each purge step, the film was exposed to an oxygen plasma. Plasma powers of 250 W and 300 W and durations of 3 seconds and 6
seconds were utilized for Hf and Zr cycles, respectively, and the chamber pressure was kept at 15 mTorr with 60 sccm of oxygen flow during both.

Fully processed film compositions were measured utilizing a PHI Versaprobe III XPS system with monochromated Al Kα radiation with an incidence angle of 45°. Spectra were collected with a 200 µm diameter spot size, 15 kV accelerating voltage, and an anode power of 50 W. Hf 4f and Zr 3d peak intensities were fit using KolXPD XPS analysis software, and compared to determine chemical compositions.

Error contributions from film density, picosecond acoustic, and film thickness measurements were propagated into elastic moduli calculations. Film density error estimates were provided by GSAS II fitting software, and were typically on the order of ~0.5 % of the fit density value. Picosecond acoustic τ delay time errors were extracted from Gaussian positional fits to each acoustic signature, as detailed in Figure S8. For film thickness error contributions, measured spatial uniformity errors of ~1.5 % were propagated in place of XRR fitting error (~0.1 %), in order to provide the most conservative error estimates. The XRR beam footprint is larger than the picosecond acoustic fs laser footprint, making it likely that the picosecond acoustic laser probes a region of film that has a thickness that may not be representative of the global film thickness probed by XRR. Thus, utilization of the known uniformity (measured by spectroscopic ellipsometry during process development on silicon wafers) provides a more conservative estimate of the error associated with the thickness across the entire film. Equations (S1)-(S3), below, represent the error propagation procedure for the elastic moduli calculations, assuming uncorrelated errors:

\[
\sigma_E = E \left( \frac{\sigma_p}{\rho} \right)^2 + 2 \left( \frac{\sigma_{\tau L}}{v_L} \right)^2 \tag{S1}
\]

\[
\sigma_{v_L} = 2 v_L \sqrt{\left( \frac{\sigma_d}{d} \right)^2 + \left( \frac{\sigma_\tau}{\tau} \right)^2} \tag{S2}
\]
\[ \sigma_\tau = \sqrt{\left(\sigma_{x_{Al/HZO}}\right)^2 + \left(\sigma_{x_{HZO/TaN}}\right)^2} \]  

Where \( \sigma_E \) is the propagated error for the elastic modulus, \( E \) is the calculated elastic modulus, \( \sigma_\rho \) is the XRR density fit error, \( \rho \) is the XRR fit density, \( \sigma_v \) is the sound velocity error, \( v_L \) is the sound velocity, \( \sigma_d \) is the film thickness error from uniformity measurements, \( d \) is the film thickness from XRR, \( \sigma_\tau \) is the delay time error, \( \tau \) is the delay time, and \( \sigma_{x_{Al/HZO}} \) and \( \sigma_{x_{HZO/TaN}} \) are the positional errors from Gaussian fits of the acoustic peaks associated with the Al/HZO and HZO/TaN interfaces.

These elastic moduli error contributions were then propagated into biaxial stress quantities calculated on the crystallized films. Other contributions to error in biaxial stress calculations included the linear intercept and slope errors from orthogonal distance regression fit of the \( d \)-spacing vs. \( \sin^2(\psi) \) data with associated \( d \)-spacing error from LIPRAS \( ^3 \) fitting. Error for the stress-free \( d \)-spacing (\( d_0 \)) was calculated using Equation (S4) with an assumption of no error in Poisson’s ratio:

\[ \sigma_{d_0} = \sqrt{\left(\sigma_m\right)^2 + \left(\sigma_b\right)^2} \]  

Where \( \sigma_m \) is the slope error and \( \sigma_b \) is the intercept error. Error associated with the biaxial stress calculation was then determined using Equation (S5):

\[ \sigma_{\sigma_{\text{biaxial}}} = \sigma_{\text{biaxial}} \sqrt{\left(\frac{\sigma_E}{E}\right)^2 + \left(\frac{\sigma_{d_0}}{d_0}\right)^2 + \left(\frac{\sigma_b}{b}\right)^2} \]  

Where \( \sigma_{\sigma_{\text{biaxial}}} \) and \( \sigma_{\text{biaxial}} \) are the calculated biaxial stress error and biaxial stress, respectively. Equation (S5) becomes the relevant error propagation expression when Equations (2) and (3) in the manuscript text are combined and simplified to Equation (S6):
\[
\sigma_{||} = \frac{E}{d_0} \cdot \frac{d_\psi - d_0}{(1+\nu) \sin^2(\psi) - 2\nu} \approx \frac{E}{d_0} \cdot \left( \frac{\partial d_\psi}{\partial \sin^2(\psi)} \right)
\]

(S6)

as described by Prevey. In this expression \( \frac{d_\psi - d_0}{(1+\nu) \sin^2(\psi) - 2\nu} \) is taken as the slope of the linear fit of the \( d \)-spacing vs. \( \sin^2(\psi) \) data.
FIG. S1. Current loops measured on HfO$_2$ (grey), Hf$_{0.69}$Zr$_{0.31}$O$_2$ (blue), Hf$_{0.54}$Zr$_{0.46}$O$_2$ (orange), Hf$_{0.21}$Zr$_{0.79}$O$_2$ (purple), and ZrO$_2$ (green) films.
FIG. S2. GIXRD patterns measured on amorphous HfO$_2$ (grey), Hf$_{0.69}$Zr$_{0.31}$O$_2$ (blue), Hf$_{0.54}$Zr$_{0.46}$O$_2$ (orange), Hf$_{0.21}$Zr$_{0.79}$O$_2$ (purple), and ZrO$_2$ (green) films.
FIG. S3. Relative permittivity (lines, left axis) and loss tangent (open points, right axis) measured on (a) HfO$_2$, (b) Hf$_{0.69}$Zr$_{0.31}$O$_2$, (c) Hf$_{0.54}$Zr$_{0.46}$O$_2$, (d) Hf$_{0.21}$Zr$_{0.79}$O$_2$, and (e) ZrO$_2$ films.
FIG. S4. XRR patterns (blue line) and associated fits (orange line) corresponding to crystalline (a) HfO$_2$, (b) Hf$_{0.69}$Zr$_{0.31}$O$_2$, (c) Hf$_{0.54}$Zr$_{0.46}$O$_2$, (d) Hf$_{0.21}$Zr$_{0.79}$O$_2$, and (e) ZrO$_2$ samples.
FIG. S5. XRR patterns (blue line) and associated fits (orange line) corresponding to amorphous (a) HfO₂, (b) Hf₀.₆₉Zr₀.₃₁O₂, (c) Hf₀.₅₄Zr₀.₄₆O₂, (d) Hf₀.₂₁Zr₀.₇₉O₂, and (e) ZrO₂ samples.
FIG. S6. Film density (orange, left axis) and thickness (blue, right axis) extracted from XRR patterns for crystallized (filled in circles) and amorphous (open squares) films with each composition.
FIG. S7. Acoustic responses measured on amorphous HfO$_2$ (grey), Hf$_{0.60}$Zr$_{0.31}$O$_2$ (blue), Hf$_{0.54}$Zr$_{0.46}$O$_2$ (orange), Hf$_{0.21}$Zr$_{0.79}$O$_2$ (purple), and ZrO$_2$ (green) films, with signatures corresponding to the Al/HZO and HZO/TaN interfaces indicated.
FIG. S8. Schematic of picosecond acoustic wave propagation through sample.
FIG. S9. Picosecond acoustic data (grey) and associated fitted background (blue), summed calculated fit (orange), Al/HZO signature Gaussian fit (purple), and HZO/TaN signature Gaussian fit (green) for the crystallized Hf$_{0.54}$Zr$_{0.46}$O$_2$ film. The positions of the acoustic signatures of the two interfaces (red lines) separated by a delay time, $\tau$ are indicated below. The composition of the film is indicated in the upper left of the panel.
**TABLE SI.** Thickness, sound velocity \((v_L)\), delay time \((\tau)\) and density \((\rho)\) for each amorphous HZO film.

<table>
<thead>
<tr>
<th>Thickness (nm)</th>
<th>HfO₂</th>
<th>Hf₀.₆₉Zr₀.₃₁O₂</th>
<th>Hf₀.₅₄Zr₀.₄₆O₂</th>
<th>Hf₀.₂₁Zr₀.₇₉O₂</th>
<th>ZrO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>20.43 ± 0.30</td>
<td>21.00 ± 0.32</td>
<td>21.05 ± 0.32</td>
<td>21.70 ± 0.33</td>
<td>22.30 ± 0.33</td>
<td></td>
</tr>
<tr>
<td>(v_L) (m s(^{-1}))</td>
<td>5022 ± 93</td>
<td>5429 ± 114</td>
<td>6206 ± 168</td>
<td>6483 ± 130</td>
<td>7386 ± 161</td>
</tr>
<tr>
<td>(\tau) (ps)</td>
<td>7.98 ± 0.09</td>
<td>7.73 ± 0.11</td>
<td>6.76 ± 0.15</td>
<td>6.69 ± 0.09</td>
<td>6.03 ± 0.10</td>
</tr>
<tr>
<td>(\rho) (g cm(^{-3}))</td>
<td>8.37 ± 0.01</td>
<td>7.70 ± 0.01</td>
<td>6.86 ± 0.01</td>
<td>6.30 ± 0.01</td>
<td>5.55 ± 0.01</td>
</tr>
</tbody>
</table>
FIG. S10. (a) 2D XRD pattern measured on the Hf$_{0.21}$Zr$_{0.79}$O$_2$ sample with indexed HZO and MgO Debye rings indicated along (b) the associated unwarped pattern with intensity line scan profiles indicated as dotted lines. Intensity line scans are shown for the (c) HZO t/o diffraction peak used for $\sin^2(\psi)$ analysis and (d) (111), (e) (200), and (f) (022) MgO diffraction peaks used for pattern unwarping, with the associated $\psi$ angle colorations identified in the legend in panel (c). Dashed lines represent the indexing for each diffraction peak.
FIG. S11. 2D XRD patterns measured on (a) HfO₂, (b) Hf₀.₅₄Zr₀.₄₆O₂, (c) Hf₀.₂₁Zr₀.₇₉O₂, and (d) ZrO₂ samples with indexed monoclinic, tetragonal, and orthorhombic Debye rings indicated. The Debye rings from the stress-free MgO powder standard are also indicated.
FIG. S12. Change in $d$-spacing with $\psi$ angle relative to film normal (filled circles, left axis) with associated linear fit (red dotted line) and normalized peak intensities (open circles, right axis) for the (200) diffraction peak of the MgO stress-free standard adhered to the (a) HfO$_2$, (b) Hf$_{0.69}$Zr$_{0.31}$O$_2$, (c) Hf$_{0.54}$Zr$_{0.46}$O$_2$, (d) Hf$_{0.21}$Zr$_{0.79}$O$_2$, and (e) ZrO$_2$ samples.
FIG. S13. Change in t/o d-spacings with ψ angle relative to film surface normal (filled circles, left axis) with associated linear fits (red dotted line) used to calculate biaxial stress magnitudes and normalized peak intensities (open circles, right axis) for the (a-b) HfO$_2$ ((111) and (111) reflections, respectively), (c) Hf$_{0.54}$Zr$_{0.46}$O$_2$, (d) Hf$_{0.21}$Zr$_{0.79}$O$_2$, and (e) ZrO$_2$ films. The calculated biaxial stress magnitudes are tabulated in the upper left of each panel along with the HZO composition.
References:

2 S. Meyer, Data Analysis for Scientists and Engineers (John Wiley & Sons Inc., 1975).