Figure S1. Energy dispersive X-ray spectroscopy (EDX) of pristine HfO$_2$ and selected Hf$_{1-x}$Ce$_x$O$_2$ nanocrystals. The detected concentrations of hafnium and cerium are listed in Table S1.
<table>
<thead>
<tr>
<th>Precursor Hf Concentration (Relative at.%)</th>
<th>Detected Hf Concentration by EDX (Relative at. %)</th>
<th>Detected Ce Concentration by EDX (Relative at. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>80</td>
<td>100</td>
<td>Not Detected</td>
</tr>
<tr>
<td>60</td>
<td>99.75</td>
<td>0.25</td>
</tr>
<tr>
<td>50</td>
<td>96.13</td>
<td>3.87</td>
</tr>
</tbody>
</table>

Table S1. Detected concentrations of hafnium and cerium by EDX analysis based on integration of the data shown in Figure S1.
Figure S2. (A) Low-magnification transmission electron microscopy image of CeO$_2$ nanocrystals. The inset shows a HRTEM image indicating the separation between the (200) lattice planes of the cubic phase. (B) X-ray diffraction pattern of CeO$_2$ nanocrystals synthesized using 2 mmol of CeCl$_3$ and 2 mmol of Ce(O‘Bu)$_4$. Reflections of cubic CeO$_2$ are indicated in red (JCPDS # 34-0394). (C) Size distribution histogram indicating the size of the CeO$_2$ nanoparticles to be ca. 1.5 ± 0.5 nm.
**HfO\(_2\) (Monoclinic) – JCPDS # 78-0050**

<table>
<thead>
<tr>
<th>atom</th>
<th>(x)</th>
<th>(y)</th>
<th>(z)</th>
<th>occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hf1</td>
<td>0.2755(2)</td>
<td>0.0397(1)</td>
<td>0.2080(2)</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>0.0739(20)</td>
<td>0.3318(17)</td>
<td>0.3466(17)</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>0.4489(20)</td>
<td>0.7582(16)</td>
<td>0.4800(22)</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space Group = \(P2_1/c\), \(a = 5.1170(1)\), \(b = 5.1754(2)\), \(c = 5.2915(2)\) \(\alpha, \gamma = 90, \beta = 99.216(2)\)

**HfO\(_2\) (Tetragonal) – From Ref. 40**

<table>
<thead>
<tr>
<th>atom</th>
<th>(x)</th>
<th>(y)</th>
<th>(Z)</th>
<th>occupancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hf1</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0</td>
</tr>
<tr>
<td>Hf2</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>1.0</td>
</tr>
<tr>
<td>O1</td>
<td>0.5</td>
<td>0.0</td>
<td>0.21282</td>
<td>1.0</td>
</tr>
<tr>
<td>O2</td>
<td>0.5</td>
<td>0.0</td>
<td>-0.28718</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>0.0</td>
<td>0.5</td>
<td>0.28718</td>
<td>1.0</td>
</tr>
<tr>
<td>O4</td>
<td>0.0</td>
<td>0.5</td>
<td>-0.21282</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Space Group = \(P4_2/nmc\), \(a, b = 3.560000\), \(c = 5.110000\), \(\alpha, \beta, \gamma = 90\)

**Table S2.** Unit cell parameters for the monoclinic and tetragonal phases of HfO\(_2\).
Figure S3. (A) HfO$_2$ tetragonal unit cell depicting the corresponding lattice planes from Figure 3D. (B) HfO$_2$ tetragonal unit cell depicting the corresponding lattice planes from Figure 3H. The unit cells were constructed using the lattice parameters given in Ref. 40.
Figure S4. Energy dispersive X-ray spectroscopy (EDX) of pristine HfO$_2$ and selected Hf$_{1-x}$La$_x$O$_2$ nanocrystals. The detected concentrations of hafnium and lanthanum are listed in Table S3.
<table>
<thead>
<tr>
<th>Precursor Hf Concentration (Relative at.%)</th>
<th>Detected Hf Concentration by EDX (Relative at. %)</th>
<th>Detected La Concentration by EDX (Relative at. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>87.5</td>
<td>100</td>
<td>Not Detected</td>
</tr>
<tr>
<td>52.5</td>
<td>96.81</td>
<td>3.19</td>
</tr>
<tr>
<td>50</td>
<td>95.99</td>
<td>4.01</td>
</tr>
</tbody>
</table>

*Table S3.* Detected concentrations of hafnium and lanthanum by EDX analysis integrated from the spectra depicted in Figure S4.
Figure S5. (A) HfO$_2$ tetragonal unit cell depicting the corresponding lattice planes from Figure 6D. (B) HfO$_2$ tetragonal unit cell depicting the corresponding lattice planes from Figure 6H. The unit cells are constructed using lattice parameters given in Ref. 40.