Supporting Information


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Table S1. Estimated compositions of the solid solution films.

<table>
<thead>
<tr>
<th>Composition</th>
<th>BFO90-STO10</th>
<th>BFO75-STO25</th>
<th>BFO50-STO50</th>
<th>BFO25-STO75</th>
</tr>
</thead>
<tbody>
<tr>
<td>BiFeO$_3$ (BFO)$^{[a]}$</td>
<td>91.8</td>
<td>76.1</td>
<td>50.7</td>
<td>25.1</td>
</tr>
<tr>
<td>SrTiO$_3$ (STO)$^{[a]}$</td>
<td>8.2</td>
<td>23.9</td>
<td>49.3</td>
<td>74.9</td>
</tr>
</tbody>
</table>

$^{[a]}$ molar ratios estimated by energy-dispersive X-ray spectroscopy (Fe, Ti)
Figure S1. Tauc plots of BiFeO$_3$ (BFO), SrTiO$_3$ (STO) and BFO:STO solid solution films.

Figure S2. Polarization curves of BiFeO$_{375}$-SrTiO$_{325}$ films on SrRuO$_3$/SrTiO$_3$ substrates in a phosphate buffer solution (0.5 M, pH 12), under chopped AM 1.5 illumination: (a) With different substrate temperatures during pulsed laser deposition. (b) With different nominal thicknesses.
**Figure S3.** $\omega$-$2\theta$ scan of BiFeO$_3$25-SrTiO$_3$75 film on SrRuO$_3$/SrTiO$_3$ substrate.

**Figure S4.** Reciprocal space map around the (103) reflections with different $\phi$ by 90° from that shown in Figure 4(b).
Figure S5. Open-circuit potentials in the dark and under illumination.

Figure S6. Current-time curve of BFO75-STO25 film/Pt kept at 0 V vs. RHE in the solution.