Supporting information for:

**Magnetic Enhancement and Low Thermal Expansion of (1-x-y)PbTiO$_3$-xBi(Ni$_{1/2}$Ti$_{1/2}$)O$_3$-yBiFeO$_3**

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The intrinsic thermal expansion behavior of the $(1-x-y)$PbTiO$_3$-$x$Bi(Ni$_{1/2}$Ti$_{1/2}$)O$_3$-$y$BiFeO$_3$ samples was determined by high temperature X-ray diffraction. Figure S1 shows the XRD patterns at different temperatures of 0.70PT-0.20BNT-0.10BF and 0.60PT-0.25BNT-0.15BF, respectively.

**Figure S1.** XRD patterns at different temperatures of (a) 0.70PT-0.20BNT-0.10BF and (b) 0.60PT-0.25BNT-0.15BF.

Figure S2 and Figure S3 show the evolutions of lattice parameters and the unit cell volumes of the PT-BNT-BF compounds with temperature. Although the evolutions of $a(b)$ and $c$ axis with temperature are anisotropic in tetragonal PT-BNT-BF solid solutions, the bulk volume change little in a broad temperature range. The intrinsic volumetric thermal expansion coefficients were listed in Table S1.

**Figure S2.** The evolution of lattice parameters with temperature of (a) 0.70PT-0.20BNT-0.10BF and (b) 0.60PT-0.25BNT-0.15BF.
**Figure S3.** The evolution of unit cell volumes with temperature of 
(1-\(x\)-\(y\))PbTiO\(_3\)-\(x\)Bi(\(\text{Ni}_{1/2}\)Ti\(_{1/2}\))O\(_3\)-\(y\)BiFeO\(_3\).

**Table S1.** Thermal expansion coefficients of (1-\(x\)-\(y\))PT-\(x\)BNT-\(y\)BF

<table>
<thead>
<tr>
<th>Composition</th>
<th>(\bar{\alpha}_V)</th>
<th>(\bar{\alpha}_l)</th>
<th>3 (\bar{\alpha}_l)</th>
<th>Temperature Range (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.70PT-0.20BNT-0.10BF</td>
<td>0.80</td>
<td>0.29</td>
<td>0.87</td>
<td>25-500</td>
</tr>
<tr>
<td>0.60PT-0.25BNT-0.15BF</td>
<td>7.25</td>
<td>2.70</td>
<td>8.10</td>
<td>25-490</td>
</tr>
</tbody>
</table>

Generally, the intrinsic thermal expansion \(\alpha_V\) and the apparent thermal expansion \(\alpha_l\) meet the formulation: \(\alpha_V \approx 3\alpha_l\) which derived from the following computations:

For \(\alpha_V = \frac{1}{V_0} \frac{dV}{dT}\), \(\alpha_l = \frac{1}{l_0} \frac{dl}{dT}\) where \(V\) is the unit cell volume, \(l\) is the one-dimensional length of the ceramic sample, \(T\) is the temperature. As mentioned above, the ceramics in the dilatometer measurement could be considered as isotropic, the thermal expansion would be isotropic as well. Thus, the apparent thermal expansion was independent of the shape of the samples. The volumetric thermal expansion certainly is proportional to the one-dimensional thermal expansion.

For regularly shaped ceramics, ignoring the defects, the \(V = k \cdot l^3\), \((k\) is a coefficient,
and will be different values for different shape).

\[ \alpha_v = \frac{1}{V_0} \frac{d(V)}{dT} = \frac{1}{k} \frac{d(kl^3)}{dT} = \frac{1}{l_0^2} \frac{d(l^3)}{dT} = \frac{1}{l_0^3} \frac{d(l^3)}{dT} \approx \frac{3}{l_0} \frac{dl}{dT} = 3 \alpha_l \quad (l_0 \approx l) \]

In another word, the apparent \( \alpha_l \) of a material could be considered accurate if its thermal expansion coefficients meet the relationship \( \alpha_v \approx 3\alpha_l \), the Our results generally accord with the relationship (See Table S1).