

## Supporting information

### **Transformation of thermal expansion from Large Volume Contraction to Nonlinear Strong Negative Thermal Expansion in $\text{PbTiO}_3\text{-Bi}(\text{Co}_{1-x}\text{Fe}_x)\text{O}_3$ Perovskites**

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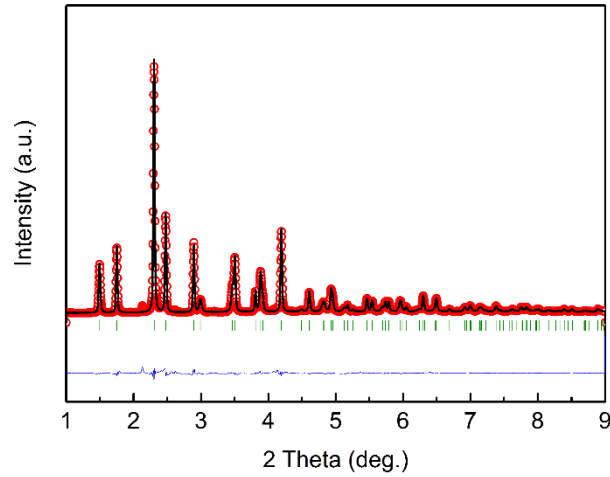
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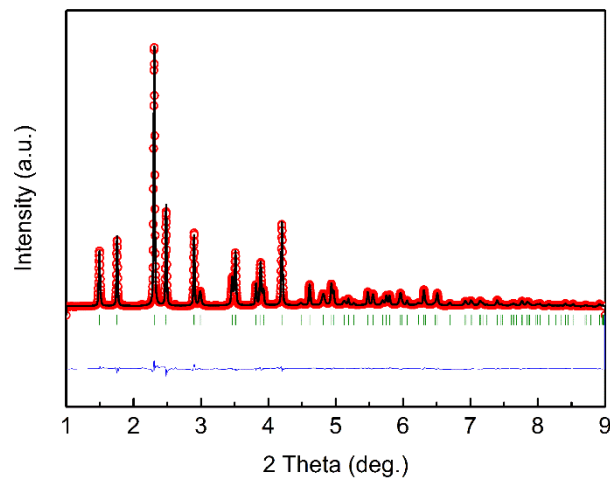
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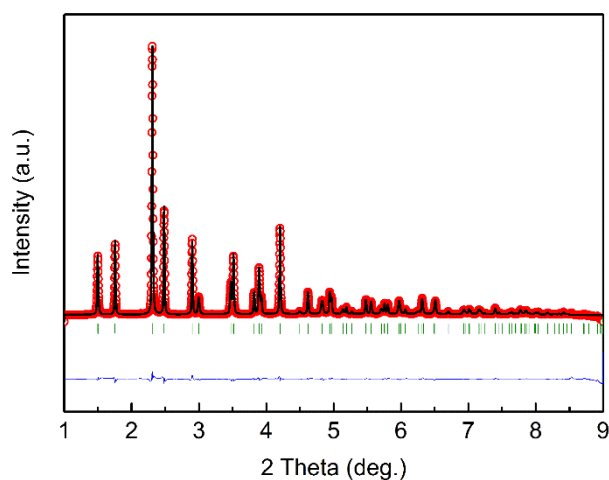
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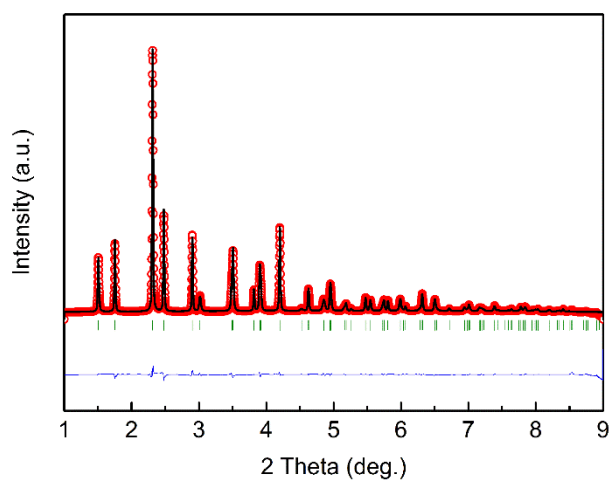
**Figure S1.** Rietveld refinement of SXRD patterns of tetragonal  $0.5\text{PbTiO}_3\text{-}0.5\text{BiCoO}_3$  at room temperature. Observed (red, circles), calculated (black line), and their difference profiles (bottom line) are shown. The Bragg reflection positions are indicated by the green ticks ( $\lambda = 0.117418 \text{ \AA}$ ).



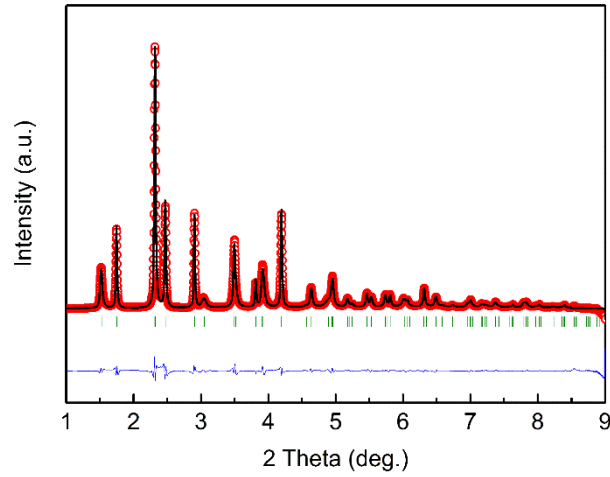
**Figure S2.** Rietveld refinement of SXRD patterns of tetragonal  $0.5\text{PbTiO}_3\text{-}0.5\text{BiCo}_{0.8}\text{Fe}_{0.2}\text{O}_3$  at room temperature. Observed (red, circles), calculated (black line), and their difference profiles (bottom line) are shown. The Bragg reflection positions are indicated by the green ticks ( $\lambda = 0.117418 \text{ \AA}$ ).



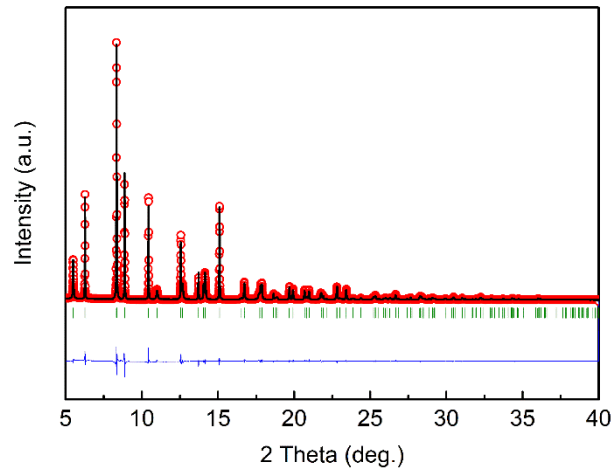
**Figure S3.** Rietveld refinement of SXRD patterns of tetragonal  $0.5\text{PbTiO}_3\text{-}0.5\text{BiCo}_{0.6}\text{Fe}_{0.4}\text{O}_3$  at room temperature. Observed (red, circles), calculated (black line), and their difference profiles (bottom line) are shown. The Bragg reflection positions are indicated by the green ticks ( $\lambda = 0.117418 \text{ \AA}$ ).



**Figure S4.** Rietveld refinement of SXRD patterns of tetragonal  $0.5\text{PbTiO}_3\text{-}0.5\text{BiCo}_{0.4}\text{Fe}_{0.6}\text{O}_3$  at room temperature. Observed (red, circles), calculated (black line), and their difference profiles (bottom line) are shown. The Bragg reflection positions are indicated by the green ticks ( $\lambda = 0.117418 \text{ \AA}$ ).



**Figure S5.** Rietveld refinement of SXRD patterns of tetragonal  $0.5\text{PbTiO}_3\text{-}0.5\text{BiCo}_{0.2}\text{Fe}_{0.8}\text{O}_3$  at room temperature. Observed (red, circles), calculated (black line), and their difference profiles (bottom line) are shown. The Bragg reflection positions are indicated by the green ticks ( $\lambda = 0.117418 \text{ \AA}$ ).



**Figure S6.** Rietveld refinement of SXRD patterns of tetragonal  $0.5\text{PbTiO}_3\text{-}0.5\text{BiFeO}_3$  at room temperature. Observed (red, circles), calculated (black line), and their difference profiles (bottom line) are shown. The Bragg reflection positions are indicated by the green ticks ( $\lambda = 0.421026 \text{ \AA}$ ).