Supporting Information

Giant polarization and high temperature monoclinic phase in a lead-free perovskite of $Bi(Zn_{0.5}Ti_{0.5})O_3$-$BiFeO_3$

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Experimental details

Polycrystalline samples of (1-*x*)Bi($Zn_{0.5}Ti_{0.5}$)O_3-*x*$BiFeO_3$ (abbreviated as (1-*x*)BZT-*x*BF, x = 0.1 ~ 0.6) were prepared by high-pressure and high-temperature synthesis using a cubic anvil-type high pressure apparatus. The raw materials of Bi_2O_3, ZnO, TiO_2, and Fe_2O_3 were stoichimetrically mixed and sealed in a gold capsule, and then treated at 6 GPa and 1373 K for 30 min. After the high pressure synthesis, the samples were carefully ground and annealed at 673 K for 1 hour and slow cooled to room temperature. The X-ray diffraction (XRD) patterns were collected with the Bruker D8 ADVANCE diffractometer for phase identification. The room temperature synchrotron XRD (SXRD) data were collected at the 11-ID-C beamline of the Advanced Photon Source with the light wavelength of 0.11715 Å. The high temperature SXRD data were collected at the BL44B2 beamline of SPring-8 with the light wavelength of 0.5 Å. All the SXRD data were analyzed by the Rietveld method with the FullProf software. The selected area electron diffraction (SAED) experiments were performed on the transmission electron microscope (TEM) with an accelerate voltages of 300 kV (Tecnai G2 F30 S-TWIN).

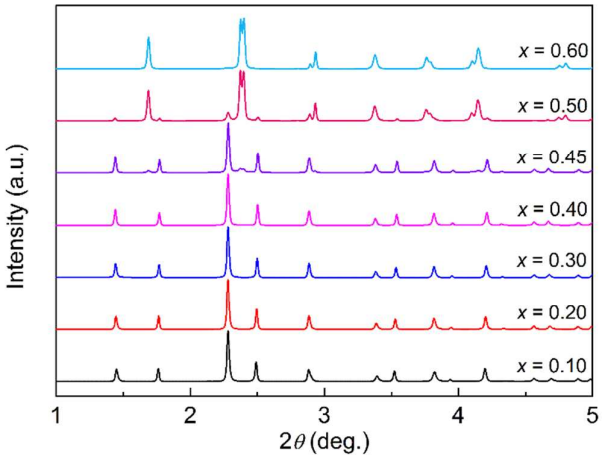

Figure S1. Synchrotron X-ray diffraction patterns (SXRD) of (1-*x*)BZT-*x*BF (x = 0.1 ~ 0.6) in a selected short range of 2θ at room temperature.

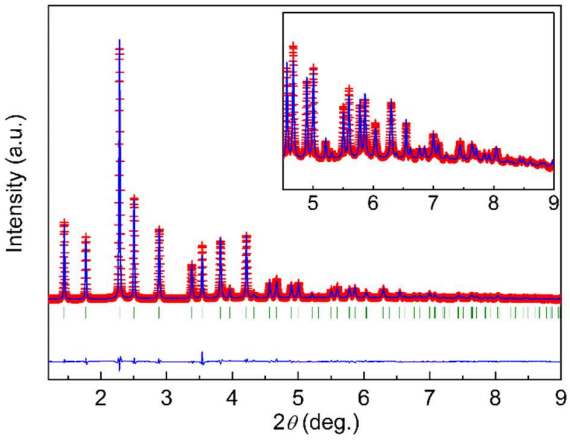

Figure S2. Rietveld full profile refinement of SXRD patterns of tetragonal 0.6BZT-0.4BF (*P*4*mm*) at room temperature. Observed (red, +), calculated (blue line), and their difference profiles (bottom line) are shown. Bragg reflection positions are indicated by the green ticks.

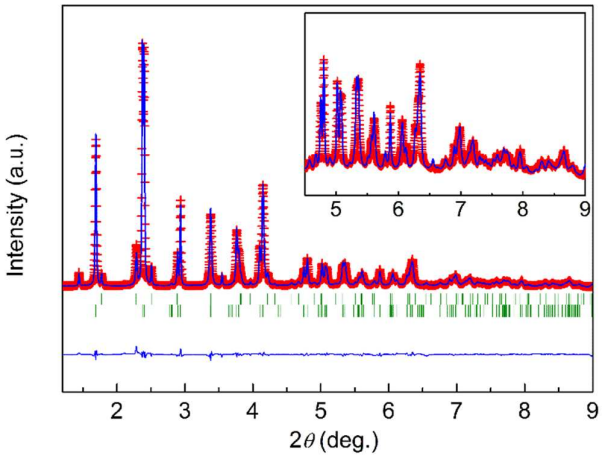

Figure S3. Rietveld full profile refinement of SXRD patterns of the MPB composition of 0.5BZT-0.5BF at room temperature. Observed (red, +), calculated (blue line), and their difference profiles (bottom line) are shown. Bragg reflection positions of the tetragonal (*P*4*mm*, upper) and monoclinic (*Cc*, lower) phases are indicated by the green ticks.

Table S1. Refined structural parameters of 0.6BZT-0.4BF with space group *P4mm* at room temperature.

Atoms	Site	g	x	y	z
Bi	1a	1.0	0	0	0
Zn	1b	0.3	0.5	0.5	0.566683(26)
Ti	1b	0.3	0.5	0.5	0.566683(26)
Fe	1b	0.4	0.5	0.5	0.566683(26)
O1	1b	1.0	0.5	0.5	0.19230(85)
O2	2c	2.0	0.5	0	0.70238(53)

*Lattice parameters: a = 3.79261 (6) Å, c = 4.65815(6) Å, $\alpha = \beta = \gamma = 90°$;

R factors (%): R_p = 9.03, R_{wp} = 8.45, R_{exp} = 2.48, χ^2 = 11.6.

Table S2. Refined structural parameters of 0.4BZT-0.6BF with space group *Cc* at room temperature.

Atoms	Site	*g*	*x*	*y*	*z*
Bi	4a	1.0	0	0.25	0
Zn	4a	0.2	0.28464(23)	0.25248(177)	0.83051(14)
Ti	4a	0.2	0.28464(23)	0.25248(177)	0.83051(14)
Fe	4a	0.6	0.28464(23)	0.25248(177)	0.83051(14)
O1	4a	1.0	0.06430(95)	0.28932(185)	0.67125(260)
O2	4a	1.0	0.34878(93)	0.48263(149)	0.14558(284)
O3	4a	1.0	0.25672(116)	0.03968(223)	0.12638(232)

*Lattice parameters: a = 9.68274(23) Å, b = 5.59094(10) Å, c = 5.64870(13) Å, β =124.86485(155)°;

R factors (%): R_p = 5.30, R_{wp} = 5.73, R_{exp} = 2.20, χ^2 = 6.78.