Supporting Information

Large magnetoelectric coupling in magnetically short-range ordered $Bi_5Ti_3FeO_{15}$ film

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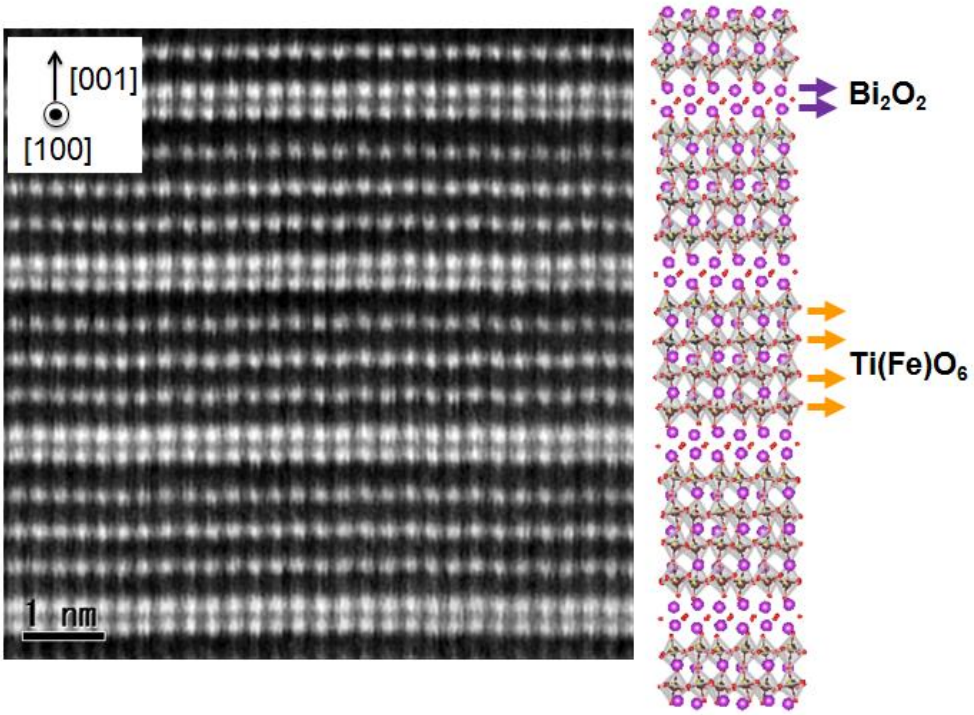

Figure S1. High-angle annular dark-field scanning TEM (HAADF STEM) picture. It further confirmed the layered structure, which shows three layers of Bi atoms $(Bi_3Ti_4O_{13})^{2-}$ sandwiched by two closely stacked Bi layers $(Bi_2O_2)^{2+}$. Between the two $(Bi_2O_2)^{2+}$ layers, there are four $Ti(Fe)O_6$ octahedra.

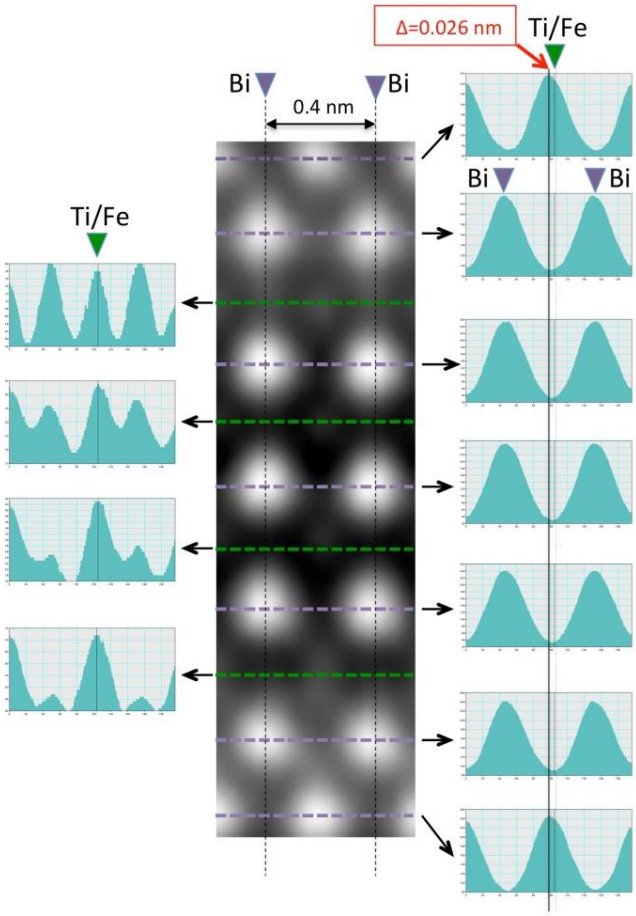

Figure S2. Atomic shift of the Ti/Fe columns, as elucidated from an image analysis of the STEM HAADF image. The shift was estimated as 0.026 nm from the centre of the two adjacent columns, which appears to be closely related to the ferroelectricity of the compound.

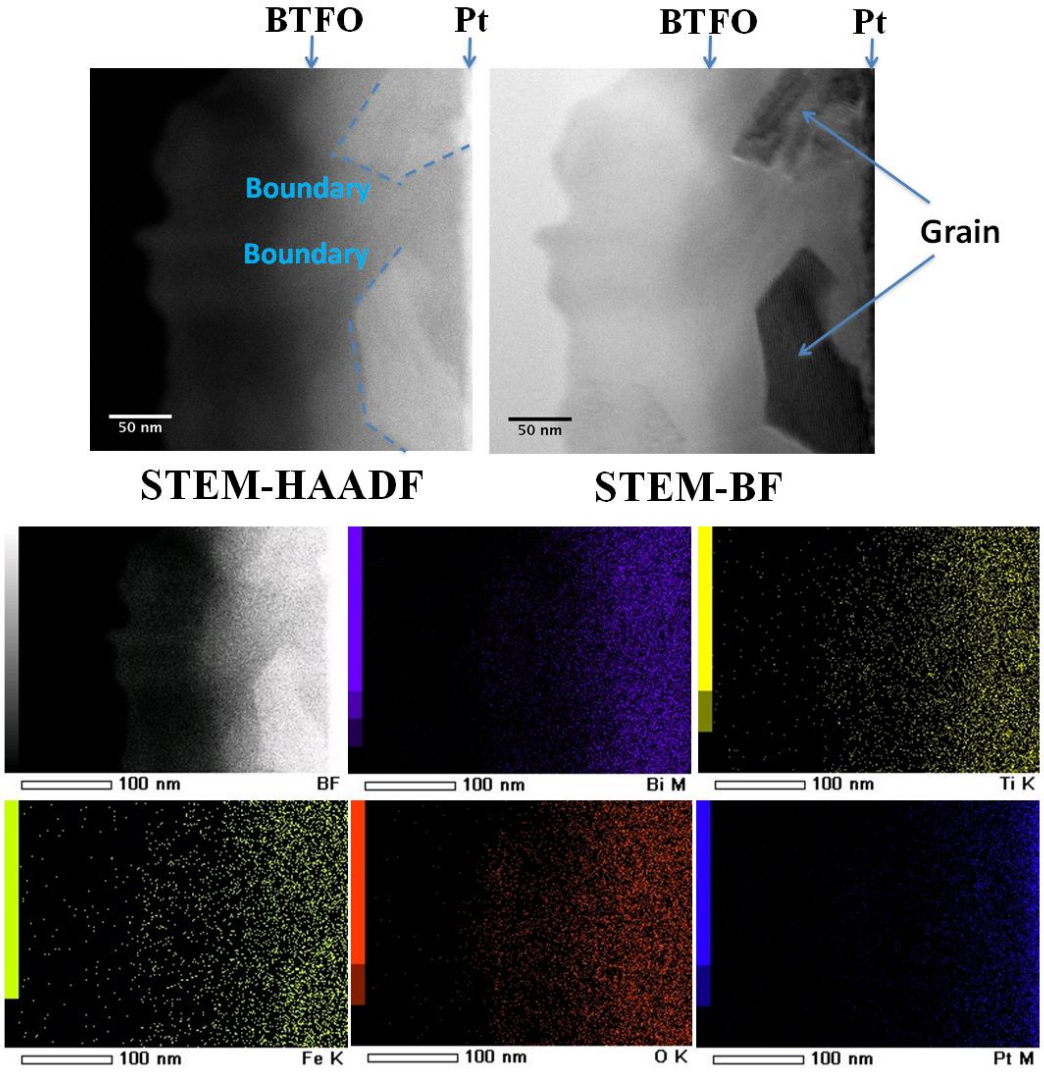

Figure S3. EDX results around the grain boundary in the BTFO film. It proved that there is no Fe aggregation at the grain boundary.

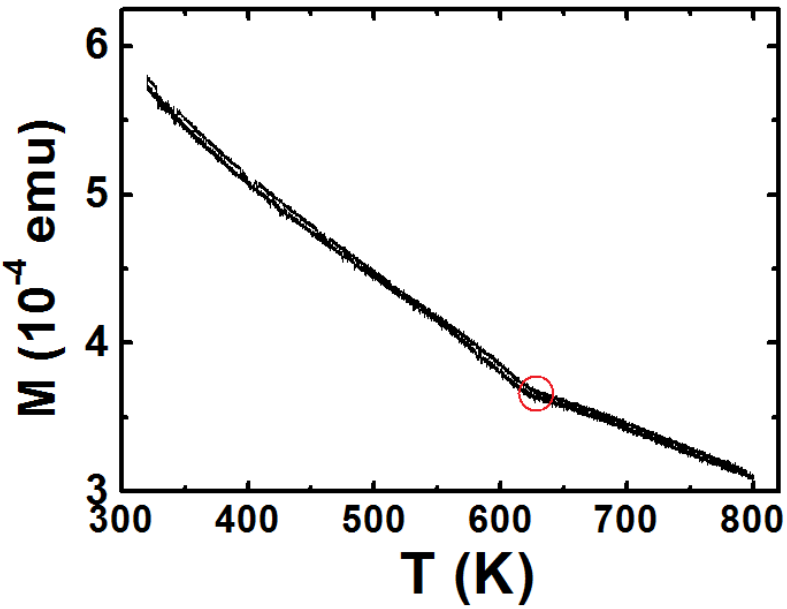

Figure S4. ZFC-FC curves measured at 1000 Oe above room temperature for ceramic sample; a transition temperature (~ 620 K), the same as for the thin film sample, was also observed.

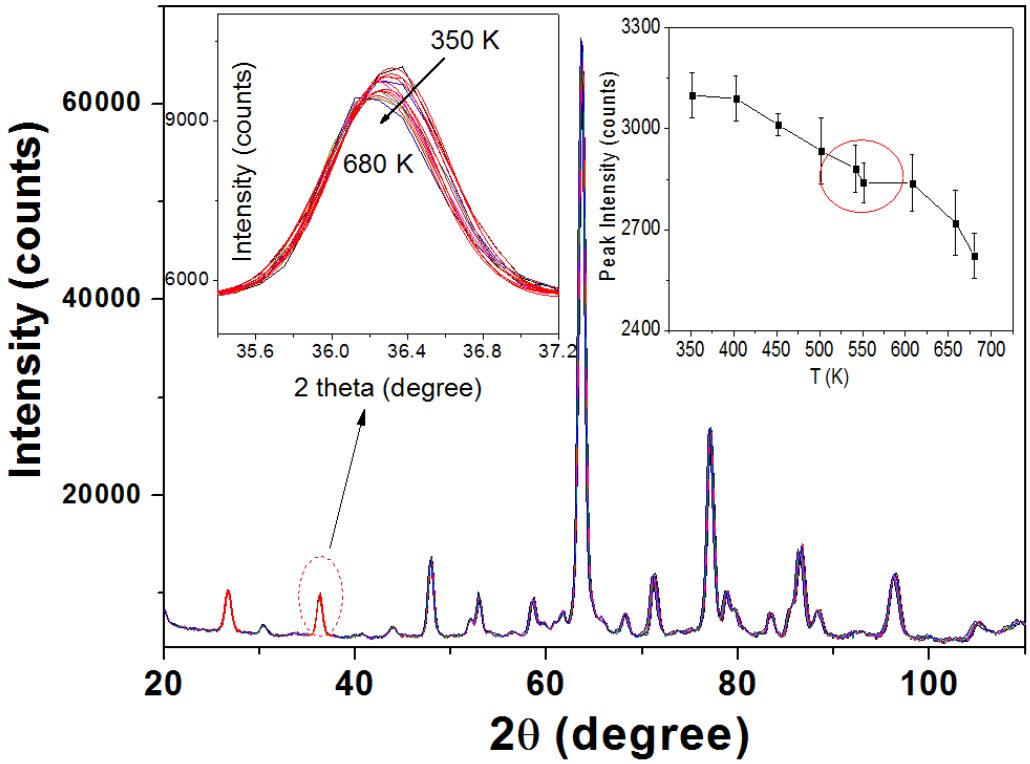

Figure S5. Neutron powder diffraction of the BTFO ceramics and analysis of a specific peak (insets). The diffraction peak intensity decreases along with temperature increase. An abnormal feature was observed around ~600 K (right inset), which is regarded due to the short-range magnetic ordering caused by spin-phonon interaction. Furthermore, no impurity peak was observed in neutron diffraction pattern of ceramics, in contrast to the XRD pattern of a thin film form. Neutron diffraction was carried out on the powder samples using the high intensity diffractometer Wombat (λ = 2.410 Å) of the OPAL Research Reactor, Lucas Heights. Over the whole temperature range (5–680 K), no magnetic peak was detected.

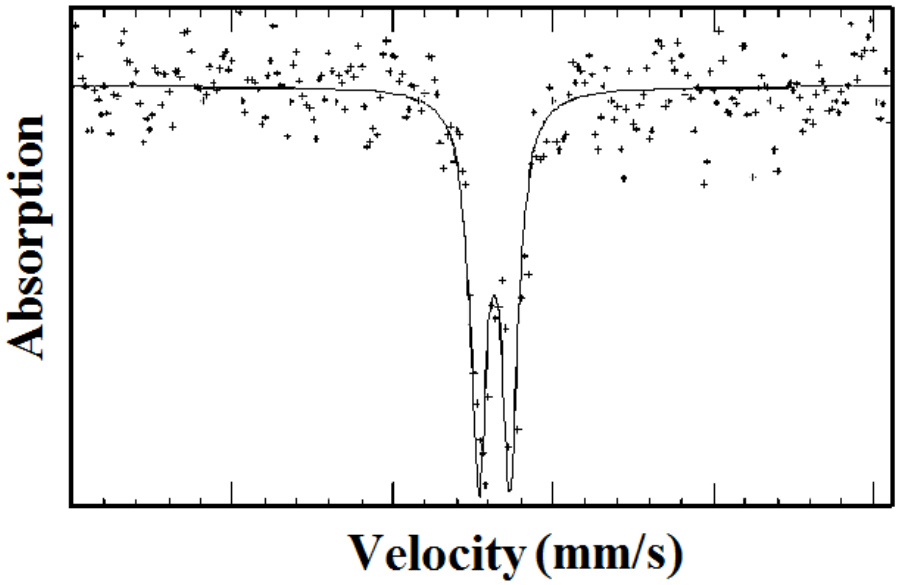

Figure S6. Mössbauer spectra obtained at room temperature using a standard constant-acceleration spectrometer and a ^{57}CoRh source. The spectra were calibrated at room temperature with a piece of α-Fe foil. Mössbauer spectra of our ceramic sample at room temperature only displayed the quadrupolar effect and can be fitted by one doublet, which indicated that there is no long-range magnetic ordering.

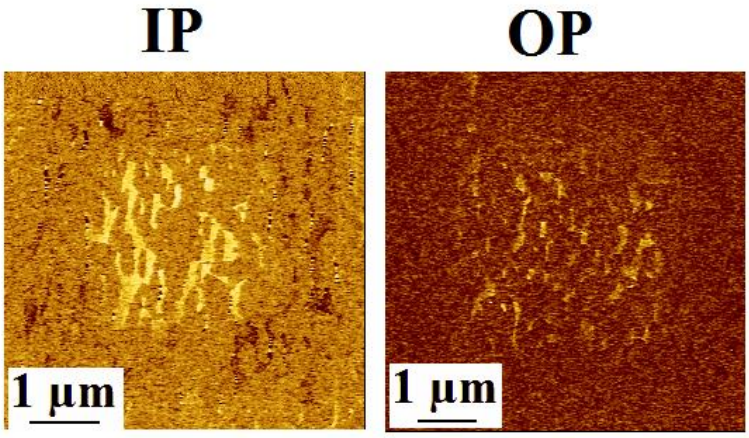

Figure S7. After applied positive and negative voltage, obvious change in the contrast was observed both in IP and OP PFM. Left: IP PFM image poled with ± 6 V; Right: OP PFM image poled with ± 10 V.

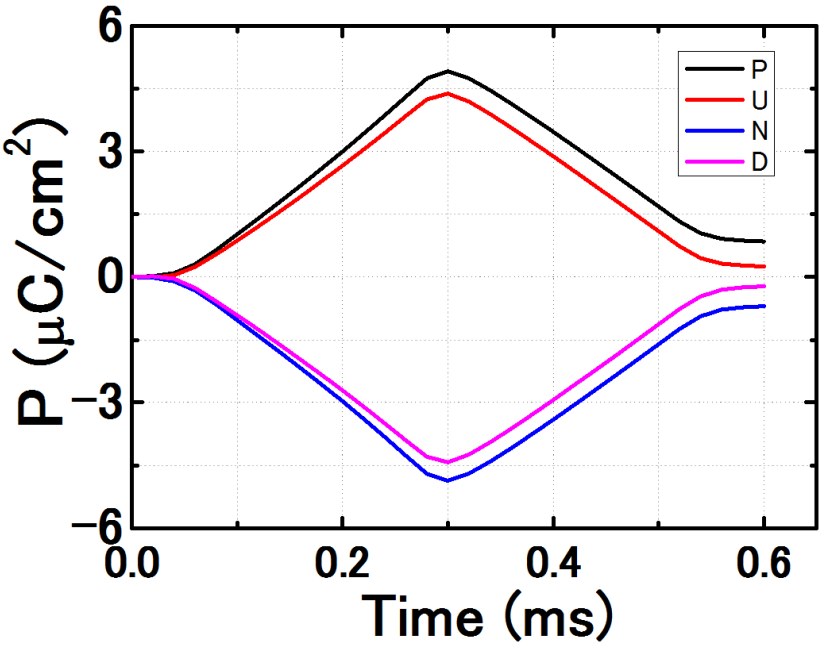

Figure S8. PUND measurement of BTFO film at room temperature. As the definition of ferroelectricity is strict, the positive-up-negative-down (PUND) measurement was done and the obvious switchable polarization was observed. The triangle voltage waveform was used, PUND amplitude is ±8V, the frequency is 1000Hz, the write pulse rise-time is 0.25ms and the read pulse delay is 0.1ms.