Supplementary Information (SI) for Journal of Materials Chemistry C.
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Supplementary material

Room temperature Multiferroicity and Magnetoelectric coupling in Ca/Mn modified $BaTiO_3$

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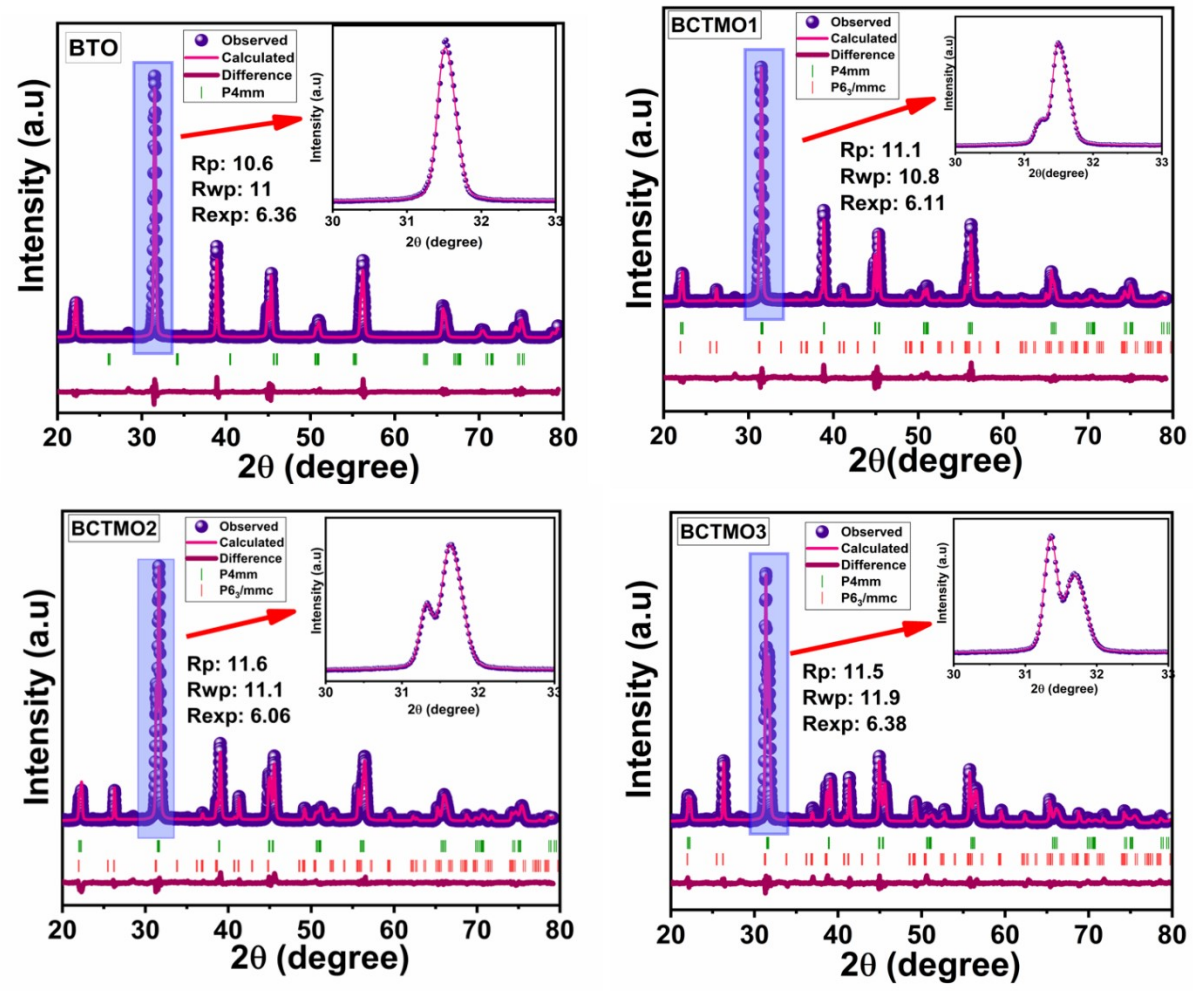

Figure S1: Refinement plots of XRD data of all samples. Inset shows the zoomed image of the emergence of hexagonal (104) peak near tetragonal (110) peak.

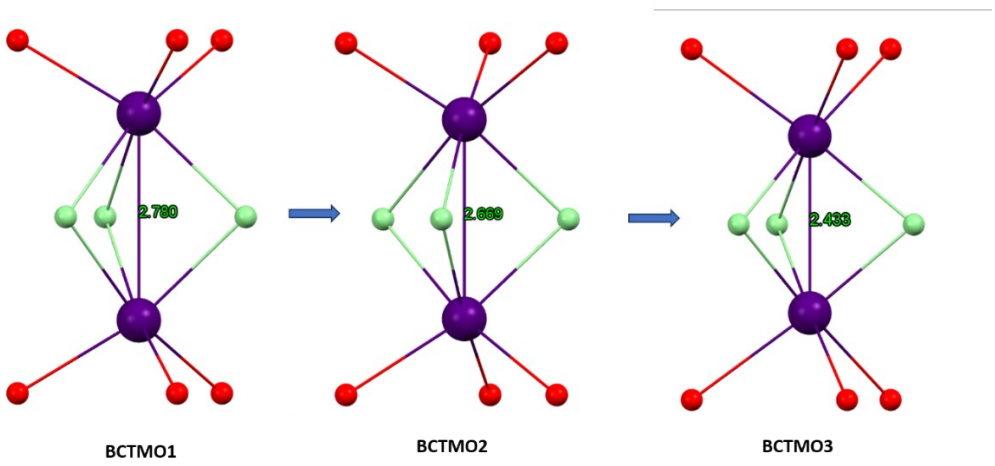

Figure S2: Variation of Ti-Ti bond in octahedral dimer of $P6_3/mmc$ space group in doped samples.

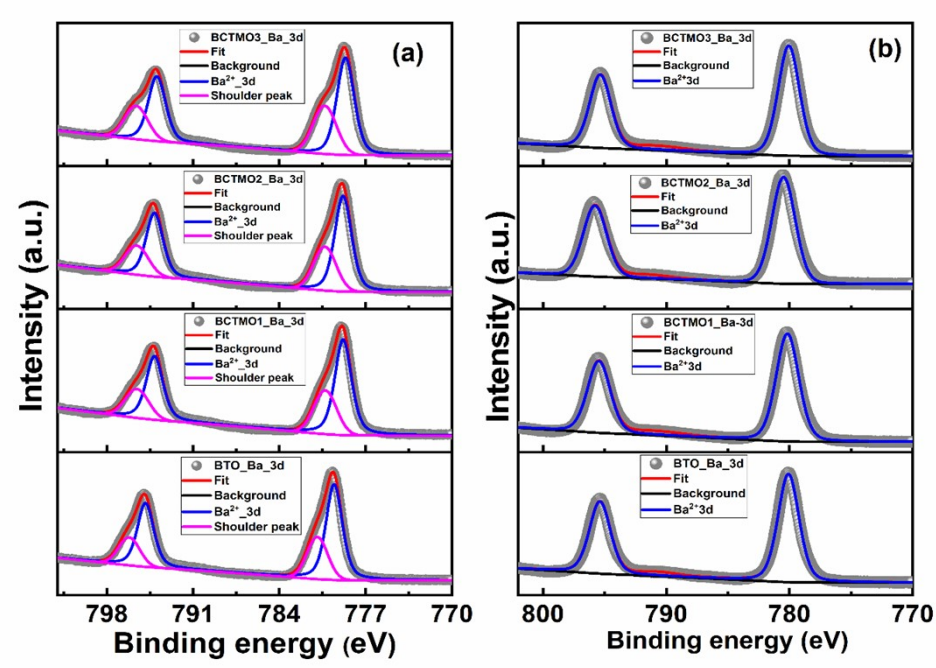

Figure S3_1: Deconvoluted Ba 3d XPS spectra **(a)** Before Ar ion etching, **(b)** After Ar ion etching

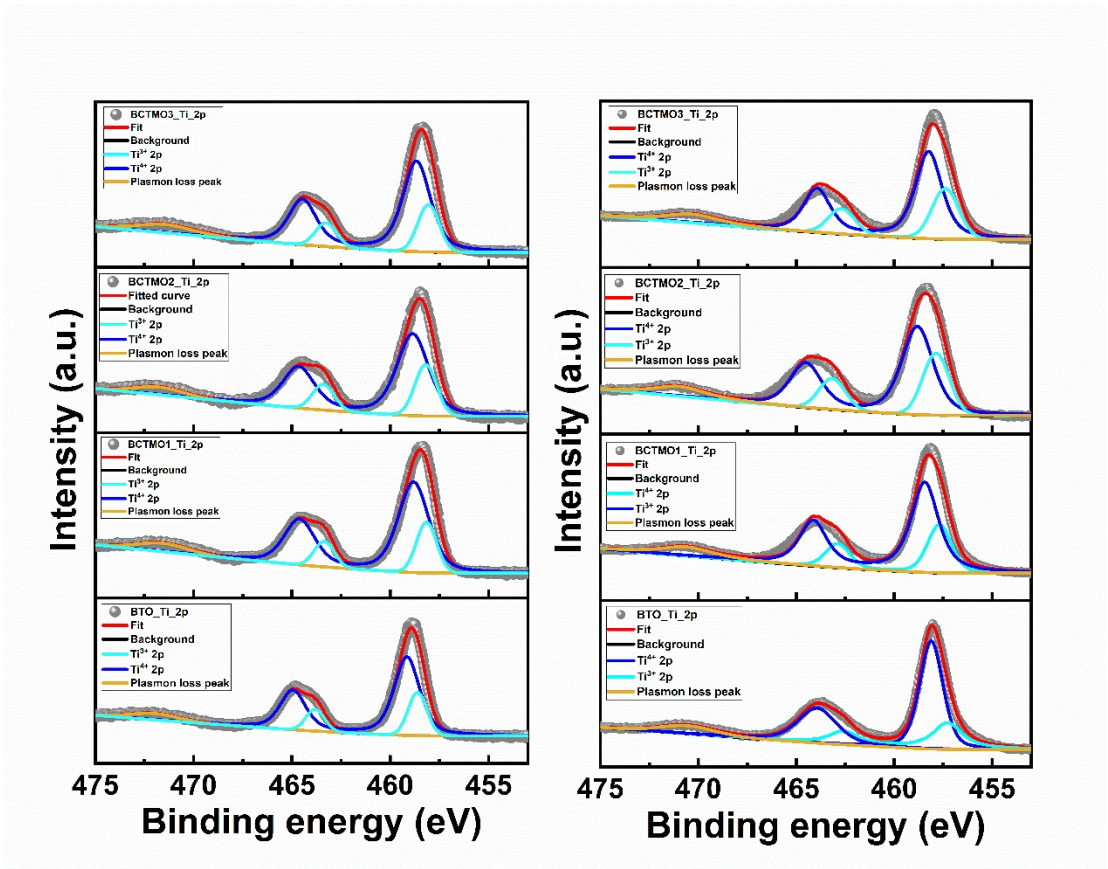

Figure S3_2: Deconvoluted Ti 2p XPS spectra **(a)** Before Ar ion etching, **(b)** After Ar ion etching

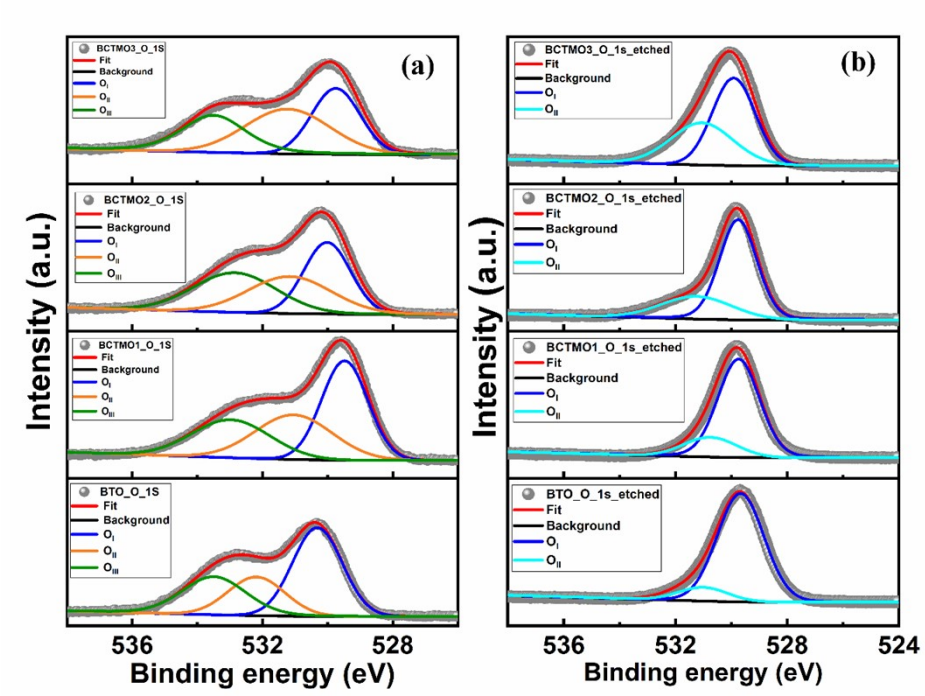

Figure S3_3: Deconvoluted O 1s XPS spectra **(a)** Before Ar ion etching, **(b)** After Ar ion etching

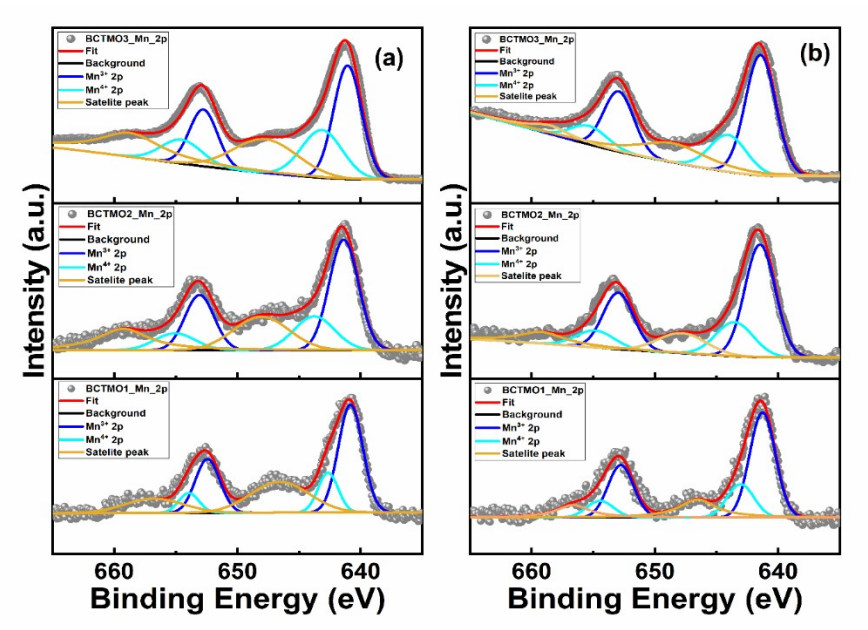

Figure S3_4: Deconvoluted Mn 2p XPS spectra **(a)** Before Ar ion etching, **(b)** After Ar ion etching

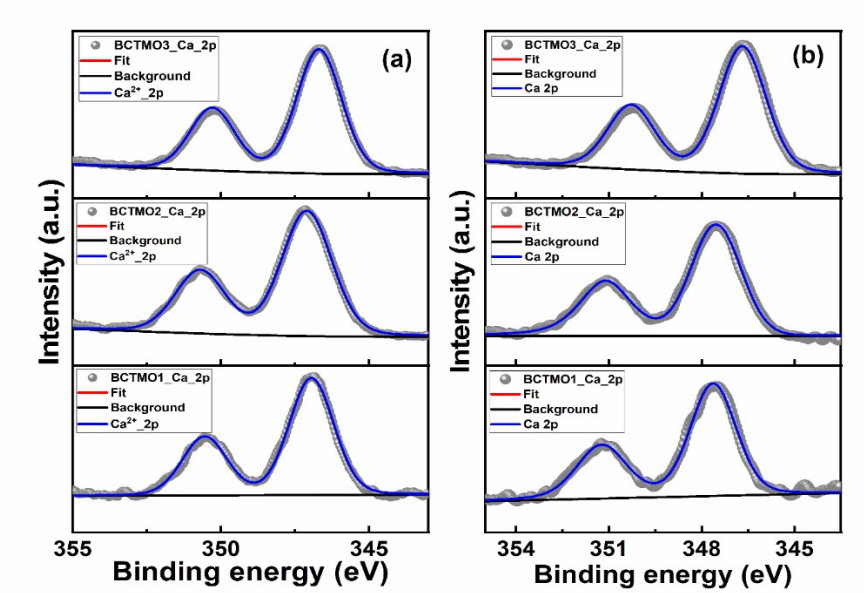

Figure S3_5: Deconvoluted Ca 2p XPS spectra **(a)** Before Ar ion etching, **(b)** After Ar ion etchin

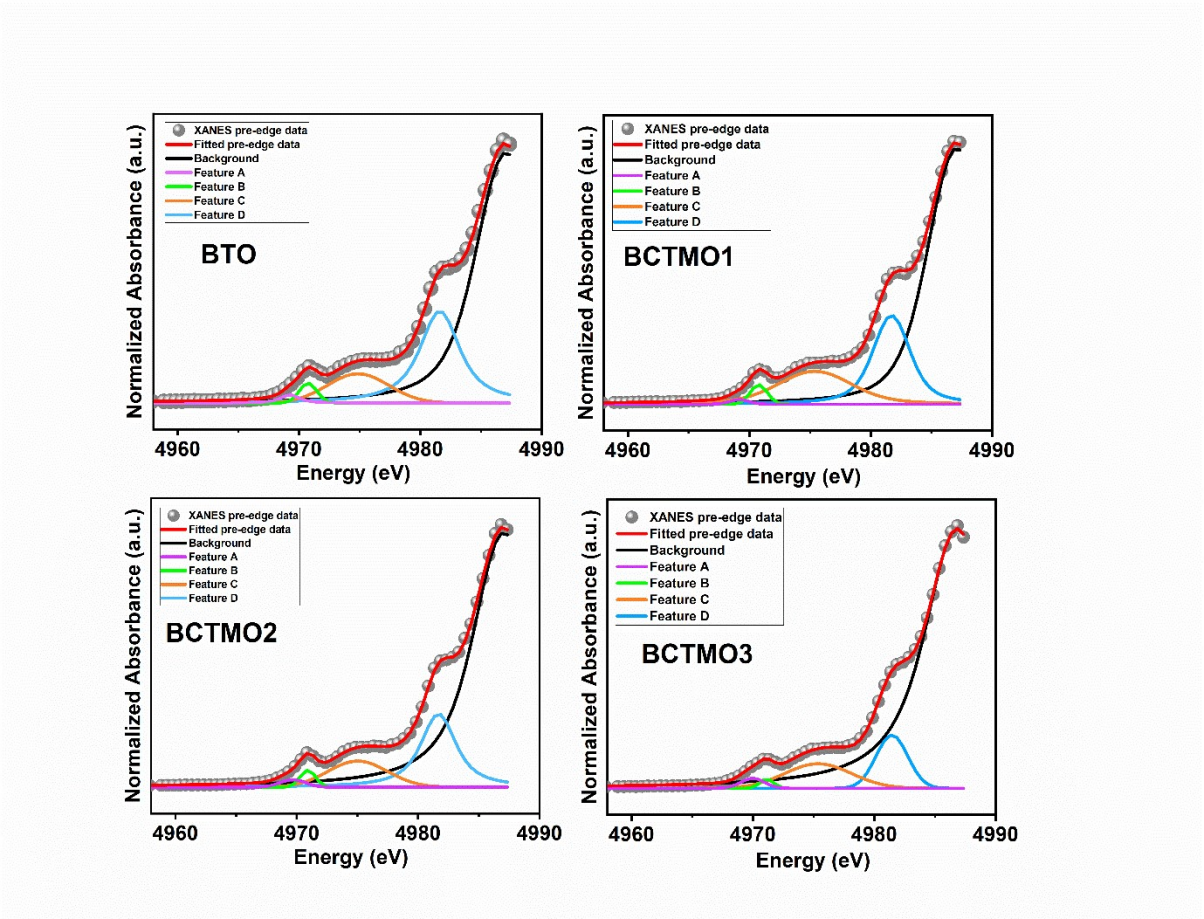

Figure S4: XANES Pre-edge fitting using Athena software

Explanation SE1: PUND measurement

The PUND measurement is a standard ferroelectric test consisting of five pulses applied in sequence. The pulses are of the same (programmable) pulse width, with a fixed delay time between the pulses and are of the same magnitude ($|V_{Max}|$). The first pulse is in the negative V_{Max} direction. It is not measured, but is used to preset the sample into the particular polarization ($\mu C/cm^2$) state. The next two pulses are in the positive V_{Max} direction. The first switches the polarization and the second does not so that both switched and unswitched polarization are measured. At each pulse measurements are made with the pulse voltage applied after the pulse width and again after the voltage returns to zero and a delay of the pulse width (ms). The last two pulses are in the negative V_{Max} direction with the first pulse switching the sample and the last pulse maintaining the switched state. During the first voltage pulse (P), all active mechanisms will bring their contribution to the measured current, including leakage currents, dielectric displacement current, and ferroelectric displacement current, During the second voltage pulse of same polarity (U), since all dipoles have been switched, leakage

currents, dielectric displacement current will contribute to this pulse. Hence, the subtraction of polarization in U from polarization in P gives the value of actual polarization.

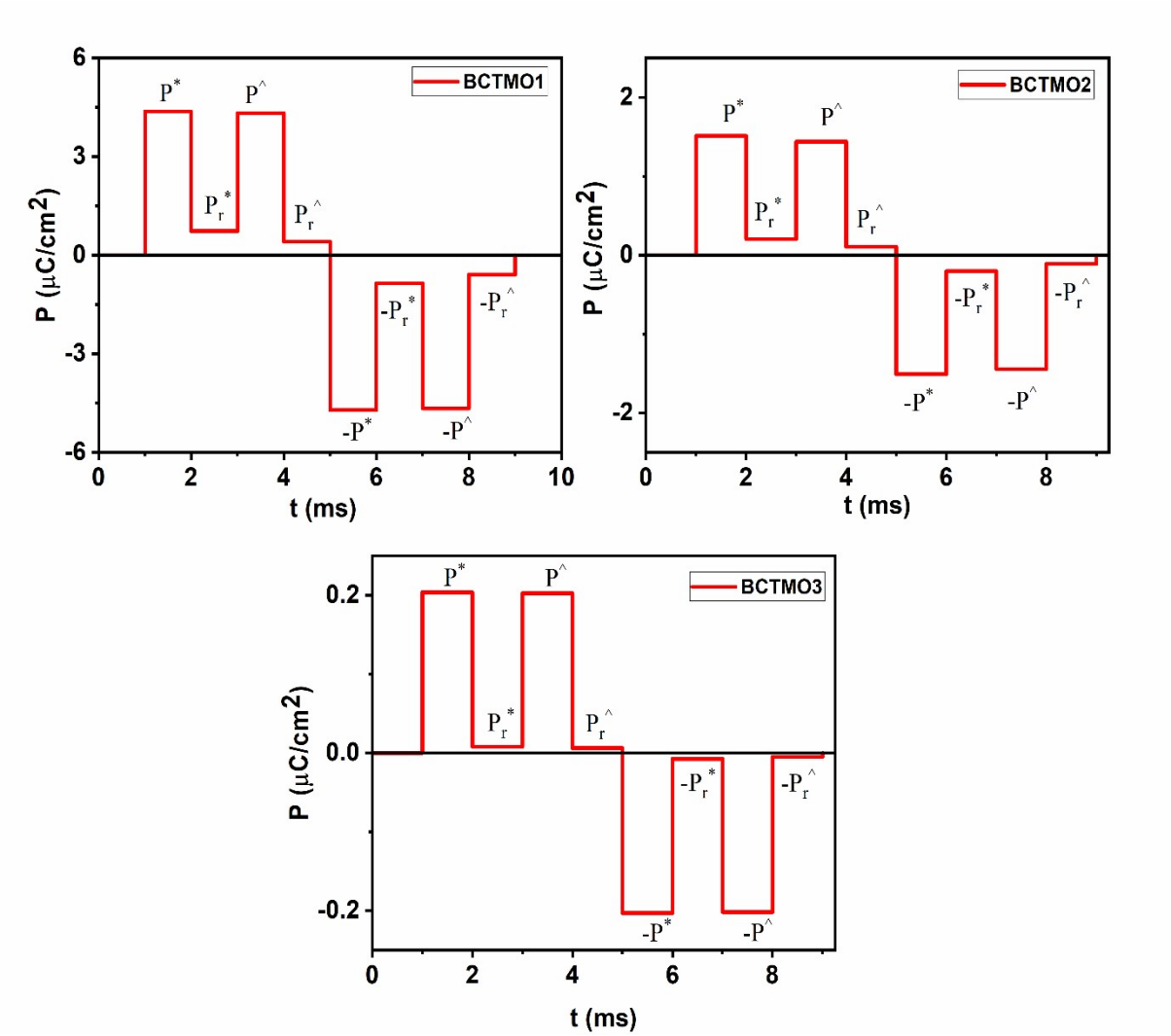

Figure S5: PUND measurement results of BCTMO samples

Table ST1:

Table ST1: PUND measurement results of the BCTMO samples with unsaturated P-E loops					
Pulse	Applied voltage (kV) (V_{max})	Measured values	BCTMO1 ($\mu C/cm^2$)	BCTMO2 ($\mu C/cm^2$)	BCTMO3 ($\mu C/cm^2$)
1	-1.5	None	-	-	-
2	1.5	P^*	4.36908	1.51114	0.20365
	0	P_r^*	0.73448	0.20373	0.00794
3	1.5	P^	4.32061	1.4952	0.20256
	0	$P_r^\wedge$	0.41005	0.10576	0.00619
4	-1.5	P^*	-4.71032	-1.50941	-0.203
	0	P_r^*	-0.8522	-0.20289	-0.00729
5	1.5	P^	-4.65976	-1.47351	-0.20169
	0	$P_r^\wedge$	-0.59091	-0.10943	-0.00501
Net Switching Polarization, dP			0.04951	0.02592	0.0012
True remnant polarization dP_r			0.2929	0.0957	0.0020