Supporting Information

Structural evolution and coexistence of ferroelectricity and antiferromagnetism in Fe, Nb co-doped $BaTiO_3$ ceramics

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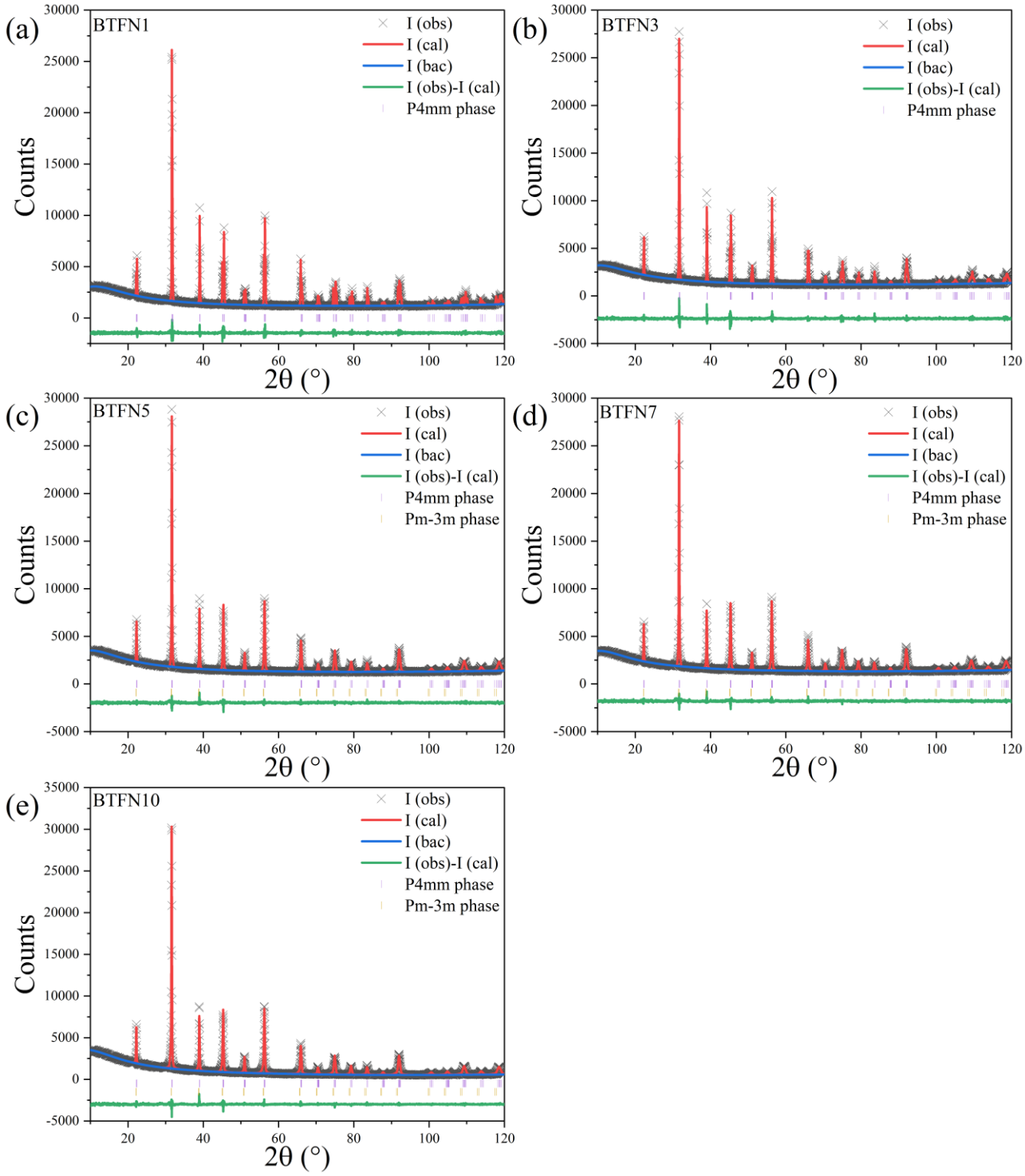

Fig. S1. Fitted X-ray powder diffraction profiles for BTFN compositions: (a) x = 0.01, (b) x = 0.03, (c) x = 0.05, (d) x = 0.07, and (e) x = 0.10. Observed (black crosses), calculated (red solid line), background (blue solid line) and difference (green solid line) profiles are shown along with the Bragg peak positions (yellow and purple markers).

Table S1. Crystal and refinement parameters for $BaTi_{1-x}(Fe_{0.5}Nb_{0.5})_xO_3$ ceramics ($0.01 \leq x \leq 0.10$).

Sample	BTFN1	BTFN3	BTFN5		BTFN7		BTFN10	
Phase	Tetragonal	Tetragonal	Tetragonal	Cubic	Tetragonal	Cubic	Tetragonal	Cubic
Space group	*P*4*mm*	*P*4*mm*	*P*4*mm*	*Pm*-3*m*	*P*4*mm*	*Pm*-3*m*	*P*4*mm*	*Pm*-3*m*
Weight fraction	100%	100%	96.6(1)%	3.4(2)%	95.8(2)%	4.2(2)%	79.4(4)%	20.6(1)%
Lattice parameters (Å)	a = 3.9994(5)	a = 4.0040(6)	a = 4.0080(1)	a = 4.0356(5)	a = 4.0101(1)	a = 4.0354(5)	a = 4.0076(1)	a = 4.0338(1)
	c = 4.0341(8)	c = 4.0257(5)	c = 4.0208(1)		c = 4.0209(1)		c = 4.0197(2)	
Volume (Å^3)	64.529(2)	64.541(3)	64.592(5)	65.726(28)	64.661(5)	65.718(28)	64.561(4)	65.637(27)
[a]R-factors	wRp = 0.0397	wRp = 0.0386	wRp = 0.0316		wRp = 0.0311		wRp = 0.0426	
	Rp = 0.0295	Rp = 0.0284	Rp = 0.0244		Rp = 0.0241		Rp = 0.0326	
	R_{exp} = 0.0244	R_{exp}= 0.0241	R_{exp} = 0.0234		R_{exp} = 0.023		R_{exp} = 0.0384	
	χ^2 = 2.655	χ^2 = 2.580	χ^2 = 1.831		χ^2 = 1.840		χ^2 = 2.260	
No. of reflections	92	92	92	44	92	44	94	44
No of variables	27	25	27		25		28	
No. of data points	3490	3490	3490		3490		3490	

[a]For definition of R-factors see Reference [1].

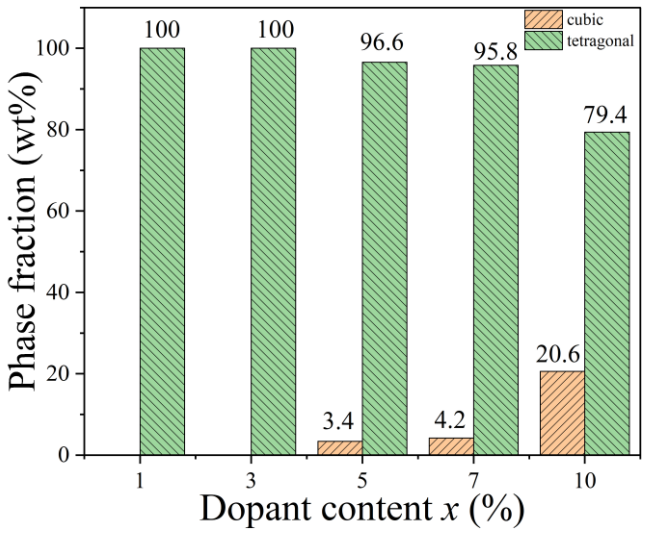

Fig. S2. Variation of phase fraction (wt%) in BTFN ceramics on doping.

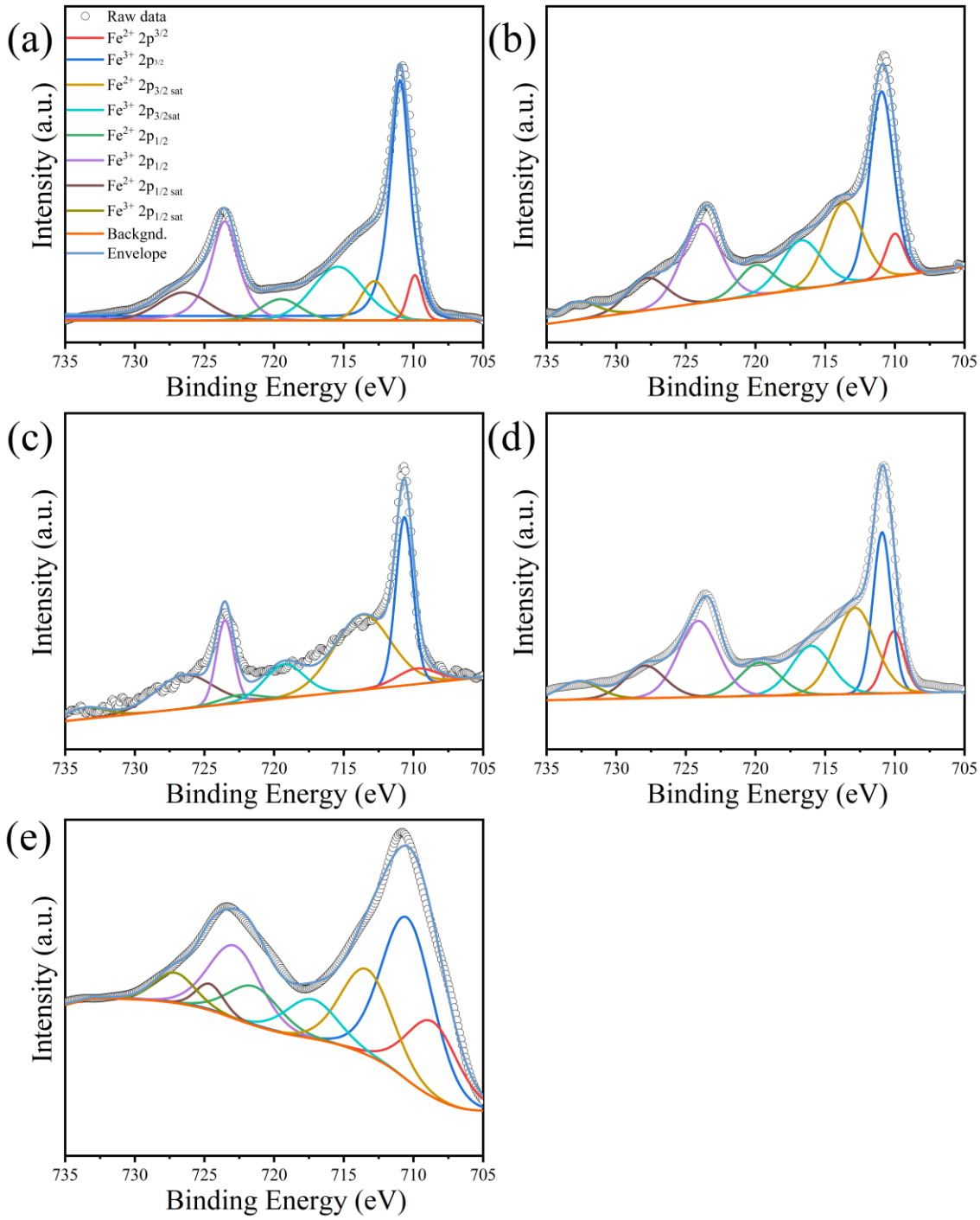

Fig. S3. Fitted Fe 2p XPS spectra for $BaTi_{1-x}(Fe_{0.5}Nb_{0.5})_xO_3$ ceramics: (a) BTFN1, (b) BTFN3, (c) BTFN5, (d) BTFN7, and (e) BTFN10.

Table S2. Binding energies and Fe^{2+}:Fe^{3+} and O_{vac}:O_{latt} ratios, as obtained from the Fe 2p and O1s XPS spectra for $BaTi_{1-x}(Fe_{0.5}Nb_{0.5})_xO_3$ ceramics.

	BTFN1	BTFN3	BTFN5	BTFN7	BTFN10
Fe^{2+} $2p_{3/2}$ (eV)	709.95	709.95	709.95	709.95	709.20
Fe^{3+} $2p_{3/2}$ (eV)	710.95	710.95	710.95	710.95	710.95
Fe^{2+}:Fe^{3+}	0.09(2)	0.18(6)	0.22(4)	0.33(2)	0.40(5)
O_{latt} (eV)	529.28	529.30	529.60	529.68	529.78
O_{vac} (eV)	530.80	530.90	531.18	531.28	531.38
O_{abs} (eV)	531.78	531.60	531.88	532.08	532.18
O_{vac}:O_{latt}	0.049(4)	0.052(2)	0.067(5)	0.078(7)	0.095(2)
Nb^{5+} $3d_{5/2}$ (eV)	207.09	207.19	206.97	207.17	207.03
Nb^{5+} $3d_{3/2}$ (eV)	209.92	209.99	209.74	209.93	209.80

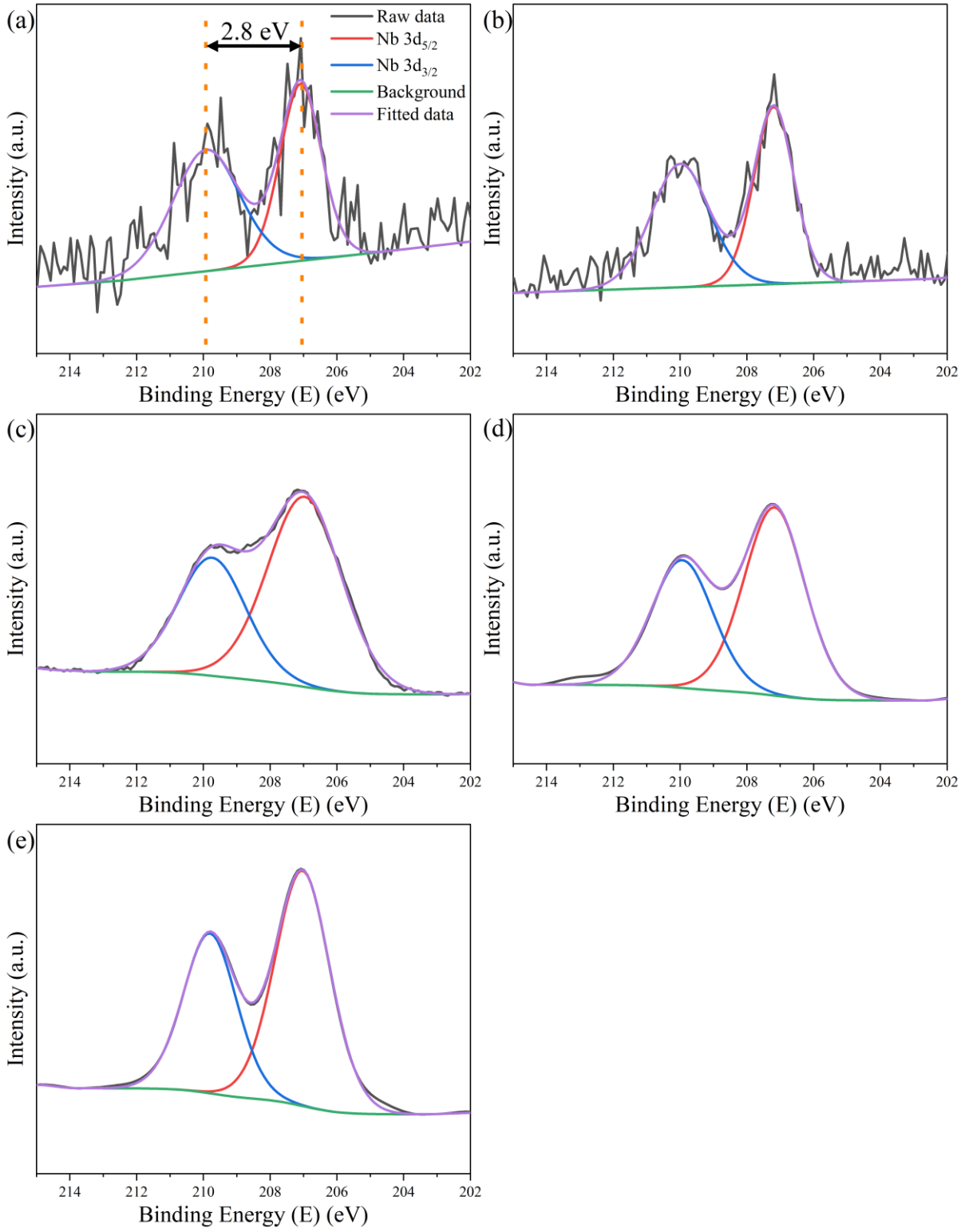

Fig. S4. Fitted Nb 3d XPS spectra for $BaTi_{1-x}(Fe_{0.5}Nb_{0.5})_xO_3$ ceramics: (a) BTFN1, (b) BTFN3, (c) BTFN5, (d) BTFN7, and (e) BTFN10.

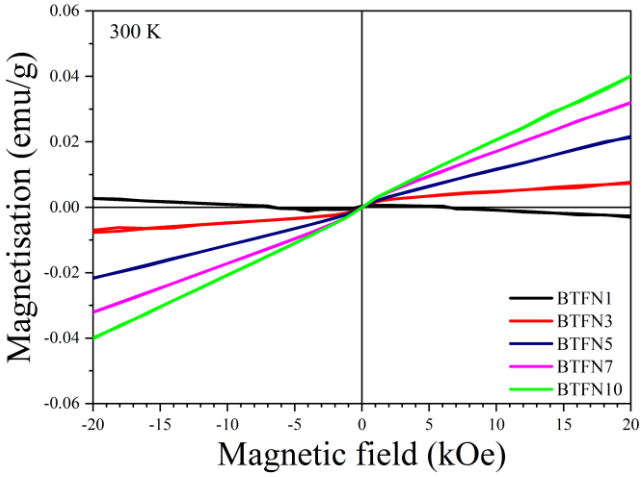

Fig. S5. *M-H* loops for $BaTi_{1-x}(Fe_{0.5}Nb_{0.5})_xO_3$ ($0.01 \leq x \leq 0.10$) ceramics at room temperature.

Reference:

[1] A.C. Larson, R. Von Dreele, Program GSAS, General Structure Analysis System, Los Alamos National Laboratories, Los Alamos. (1994).