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Supplementary Information

Unusual Crystal Structure Evolution, Multiple Phase Boundaries and Phase Coexistence in (1-x)Ba($Cu_{1/3}Nb_{2/3}$)O_3-(x)$PbTiO_3$ Perovskite Solid Solution

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Section 1

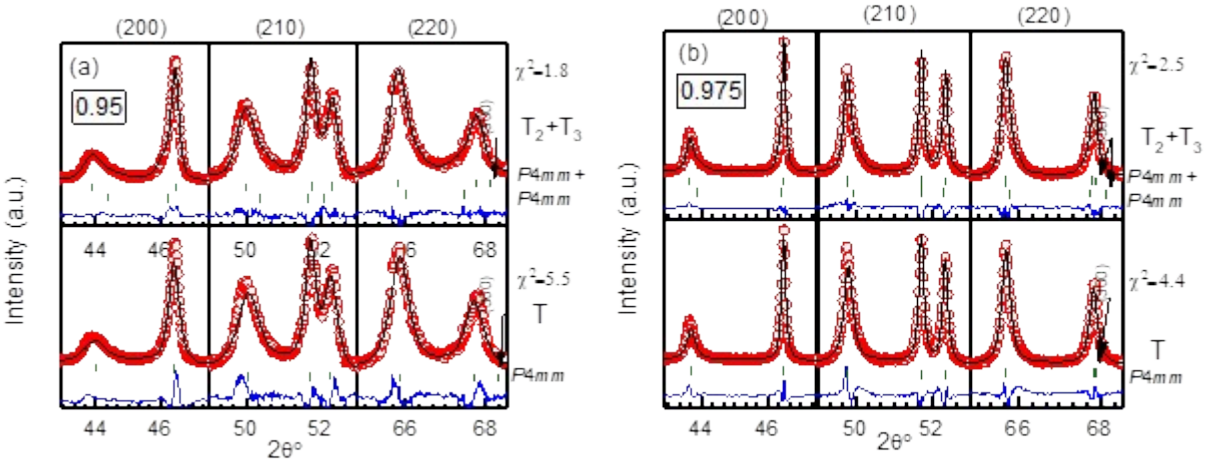

Fig. S1. Comparison of Rietveld fit of HR-XRD pattern using two coexisting tetragonal structure (upper panel) and single tetragonal structure (lower panel) for composition (a) 0.95 and (b) 0.975. The representation of Red circles, Black and Blue lines and Green ticks are observed diffraction patterns, calculated diffraction patterns, the difference between calculated and observed diffraction patterns and Bragg's reflections, respectively.

PTO

Section 2

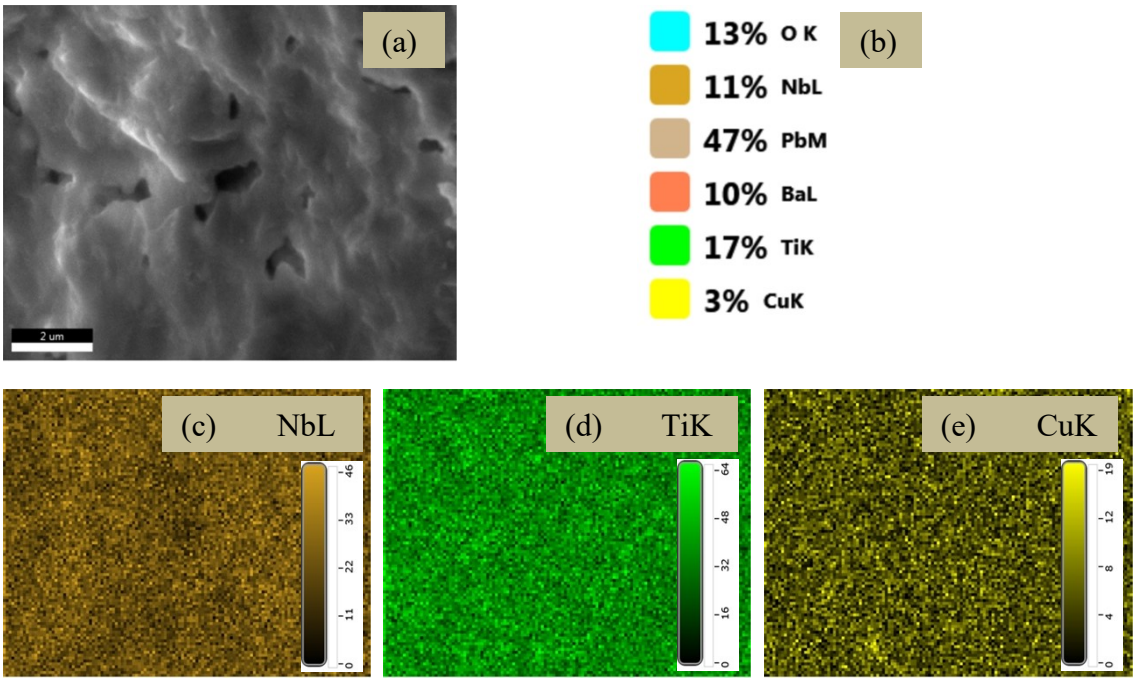

Fig. S2 (a) SEM micrograph of cross-section place where EDS imaging is taken, (b) Atomic weight percentage of constituent elements, B-site elemental mapping for (c) Nb, (d)Ti and (e) Cu-ion.

Section 3

Table: S1 Refined crystal structural parameters for composition x = 0.55.

Crystal System for 0.55		Cubic (C_1)			
Lattice Parameter (Å)	a	4.0167(2)			
Unit Cell Vol. (Å^3)		65.800(9)			
Phase fraction (%)		100			
Position Coordinates		x	y	z	B
$X_{(Ba/Pb)}$		0.0	0.0	0.0	2.14(4)
$X_{(Cu/Nb/Ti)}$		0.5	0.5	0.5	0.42(6)
X_O		0.5	0.5	0	1.3(2)
R_{wp}		8.04			
R_p		6.14			
χ^2		1.97			

Table: S2 Refined crystal structural parameters for composition x = 0.60.

Crystal System for 0.60		Cubic (C_1)				Tetragonal (T_2)			
Lattice Parameter (Å)	a; c	4.0144(1)				3.9882(1)); 4.0632(2)			
Unit Cell Vol. (Å^3)		64.697(1)				64.630(4)			
Phase fraction (%)		48.88				51.12			
Position Coordinates		x	y	z	B	x	y	z	B
$X_{(Ba/Pb)}$		0.0	0.0	0.0	2.13(5)	0.0	0.0	0.0	0.54(2)
$X_{(Cu/Nb/Ti)}$		0.5	0.5	0.5	0.21(6)	0.5	0.5	0.502(7)	0.31(1)
X_{O1}		0.5	0.5	0	0.7(2)	0.5	0.5	0.075(5)	1.43(5)
X_{O2}		-	-	-	-	0.5	0.0	0.532(6)	2.5(7)
R_{wp}		7.92							
R_p		6.05							
χ^2		1.92							

Table: S3 Refined crystal structural parameters for composition x = 0.62 [43].

Crystal System for 0.62		Tetragonal (T_1)				Tetragonal (T_2)			
Lattice Parameter (Å)	a; c	3.9894(1); 4.0502(3)				3.9807(1); 4.0694(1)			
Unit Cell Vol. (Å^3)		64.461(7)				64.486(3)			
Phase fraction (%)		52.82				45.18			
Position Coordinates		x	y	z	B	x	y	z	B
$X_{(Ba/Pb)}$		0.0	0.0	0.0	1.46(3)	0.0	0.0	0.0	1.16(3)
$X_{(Cu/Nb/Ti)}$		0.5	0.5	0.510(2)	0.33(4)	0.5	0.5	0.511(3)	0.08(5)
X_{O1}		0.5	0.5	0.052(6)	0.04(3)	0.5	0.5	0.078(4)	1.55(4)
X_{O2}		0.5	0.0	0.542(8)	3.1(3)	0.5	0.0	0.529(7)	0.25(1)
R_{wp}		8.37							
R_p		6.64							
χ^2		1.89							

Table: S4 Refined crystal structural parameters for composition x = 0.65.

Crystal System for 0.65		Monoclinic (M_1)				Tetragonal (T_2)			
Lattice Parameter (Å)	a; c	3.9854(4); 3.987(4); 4.0325(3); 90.362(4)				3.9762(1); 4.0793(2)			
Unit Cell Vol. (Å^3)		64.073(9)				64.497(4)			
Phase fraction(%)		67.45				32.55			
Position Coordinates		x	y	z	B	x	y	z	B
$X_{(Ba/Pb)}$		0.0	0.0	0.0	1.67(2)	0.0	0.0	0.0	1.30(3)
$X_{(Cu/Nb/Ti)}$		0.544(2)	0.5	0.529(2)	0.08(5)	0.5	0.5	0.517(2)	0.08(2)
X_{O1}		0.49(2)	0.5	0.012(9)	0.38(4)	0.5	0.5	0.088(4)	0.81(5)
X_{O2}		0.51(1)	0.0	0.450(5)	0.52(1)	0.5	0.0	0.504(6)	0.42(7)
X_{O3}		0.153(3)	0.5	0.478(8)	1.8(5)	-	-	-	-
R_{wp}		6.26							
R_p		4.72							
χ^2		1.46							

Table: S5 Refined crystal structural parameters for composition x = 0.85.

Crystal System for 0.85		Tetragonal (T_2)				Monoclinic (M_3)			
Lattice Parameter (Å)	a; c	3.9639(1); 4.0748(2)				3.9362(4); 3.9195(2); 4.0611(2); 90.193(8)			
Unit Cell Vol. (Å^3)		64.034(3)				62.655(8)			
Phase fraction (%)		50.07				49.93			
Position Coordinates		x	y	z	B	x	y	z	B
$X_{(Ba/Pb)}$		0.0	0.0	0.0	1.15(2)	0.0	0.0	0.0	0.51(3)
$X_{(Cu/Nb/Ti)}$		0.5	0.5	0.499(7)	0.04(3)	0.527(2)	0.5	0.514(3)	0.11(2)
X_{O1}		0.5	0.5	0.101(3)	0.82(3)	0.51(13)	0.5	0.420(8)	1.21(2)

X_{O2}	0.5	0.0	0.558(3)	0.98(4)	0.072(6)	0.5	0.36(2)	1.41(2)
X_{O3}	-	-	-	-	0.52(15)	0.5	-0.109(6)	3.43(9)
R_{wp}	7.04							
R_p	5.33							
χ^2	1.54							

Table: S6 Refined crystal structural parameters for composition x=0.90.

Crystal System for 0.90		Tetragonal (T_2)				Tetragonal (T_3)			
Lattice Parameter (Å)	a; c	3.9089(1); 4.1190(2)				3.9355(2); 4.0786(3)			
Unit Cell Vol. (Å^3)		62.940 (4)				63.172 (7)			
Phase fraction(%)		28.36				71.64			
Position Coordinates		x	y	z	B	x	y	z	B
$X_{(Ba/Pb)}$		0.0	0.0	0.0	0.10(3)	0.0	0.0	0.0	0.68(3)
$X_{(Cu/Nb/Ti)}$		0.5	0.5	0.531(2)	0.08(4)	0.5	0.5	0.494(4)	0.55(5)
X_{O1}		0.5	0.5	-0.100(5)	1.18(4)	0.5	0.5	-0.122(3)	0.85(8)
X_{O2}		0.5	0.0	0.587(3)	1.61(3)	0.5	0.0	0.578(3)	1.02(5)
R_{wp}		6.67							
R_p		5.07							
χ^2		1.83							