

Supplementary Information

Re-entrant Relaxor Ferroelectric Behaviour in Nb-Doped BiFeO₃-BaTiO₃ Ceramics

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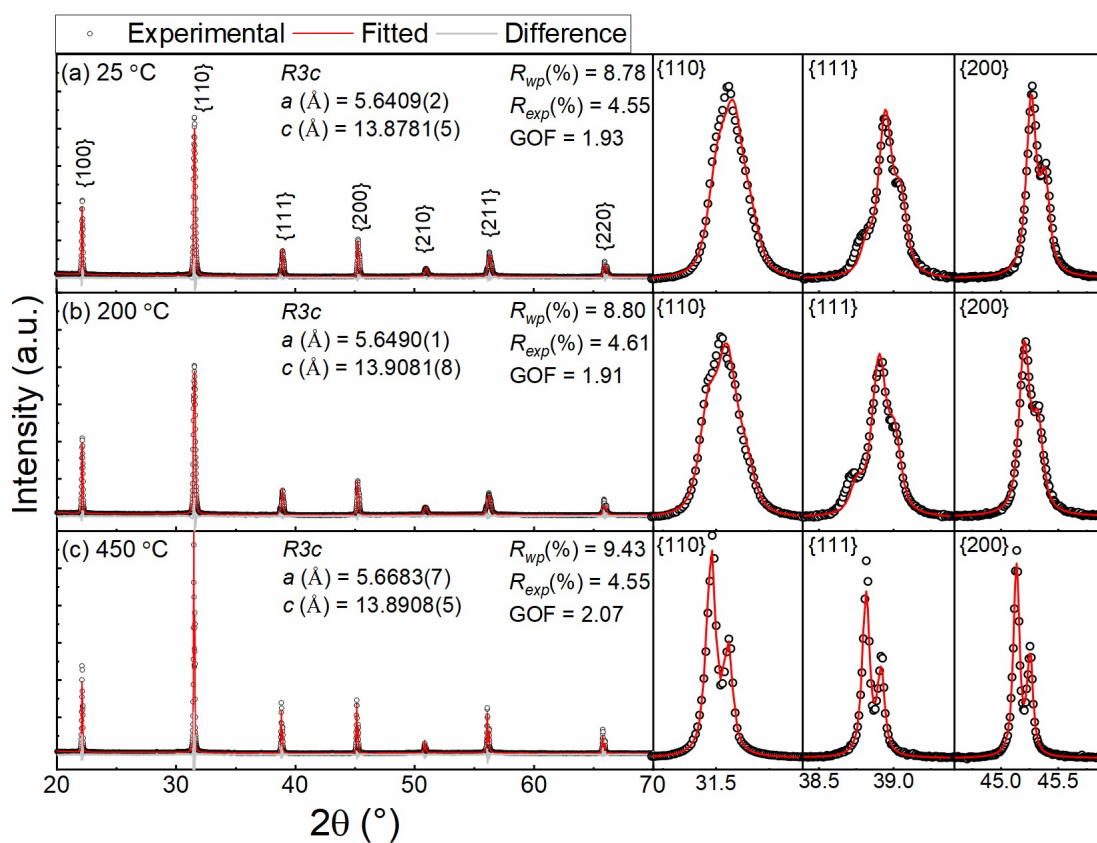


Figure S1. XRD patterns and refinements for Nb-doped 0.7BF-0.3BT ceramics at temperatures of (a) 25 °C, (b) 200 °C and (c) 450 °C respectively.

Table S1. Structural parameters of Nb-doped 0.7BF-0.3BT ceramics at different temperatures obtained from Rietveld refinement.

T (°C)	a (Å)	c (Å)	α_R (°)	α_{pc} (°)	90-α_{pc} (°)	V_R (Å³)	V_{pc} (Å³)
25	5.6409(2)	13.8781(5)	59.806(5)	89.832(5)	0.167(5)	382.42(8)	63.73(8)
50	5.6422(5)	13.8842(6)	59.797(5)	89.824(7)	0.175(3)	382.77(6)	63.79(6)
75	5.6430(4)	13.8888(5)	59.789(1)	89.817(3)	0.182(7)	383.01(1)	63.83(5)
100	5.6446(1)	13.8958(1)	59.779(3)	89.808(8)	0.191(2)	383.41(4)	63.90(2)
125	5.6453(3)	13.898(10)	59.778(1)	89.807(7)	0.192(3)	383.57(3)	63.92(9)
150	5.6466(1)	13.9020(4)	59.775(1)	89.805(2)	0.194(8)	383.85(7)	63.97(6)
175	5.6481(6)	13.9064(8)	59.773(2)	89.803(6)	0.196(4)	384.19(3)	64.03(2)
200	5.6490(1)	13.9081(8)	59.774(4)	89.804(6)	0.195(4)	384.35(5)	64.05(9)
225	5.6502(9)	13.9105(1)	59.777(1)	89.807(1)	0.193(1)	384.59(4)	64.09(9)
250	5.6521(2)	13.9136(7)	59.781(3)	89.810(6)	0.189(4)	384.93(1)	64.15(5)
275	5.6532(5)	13.9157(6)	59.783(5)	89.812(5)	0.187(5)	385.14(2)	64.19(1)
300	5.6547(9)	13.9166(7)	59.792(6)	89.820(4)	0.179(6)	385.37(8)	64.23(1)
325	5.6563(2)	13.9179(1)	59.800(6)	89.827(3)	0.172(7)	385.62(1)	64.27(1)
350	5.6576(5)	13.9168(8)	59.814(2)	89.839(1)	0.160(9)	385.77(3)	64.29(5)
375	5.6595(5)	13.9174(2)	59.827(3)	89.850(4)	0.149(6)	386.04(7)	64.34(1)
400	5.6618(6)	13.9151(1)	59.852(6)	89.872(4)	0.127(6)	386.29(8)	64.38(3)
425	5.6645(4)	13.9095(3)	59.891(2)	89.905(8)	0.094(2)	386.50(9)	64.41(8)
450	5.6683(7)	13.8908(5)	59.980(2)	89.982(8)	0.017(2)	386.51(2)	64.41(9)
400	5.6631(8)	13.9090(9)	59.881(9)	89.897(7)	0.102(3)	386.31(1)	64.38(5)
350	5.6585(1)	13.9153(9)	59.825(6)	89.849(1)	0.151(1)	385.84(8)	64.30(8)
300	5.6547(1)	13.9156(1)	59.795(3)	89.822(7)	0.177(3)	385.33(6)	64.22(3)
250	5.6514(9)	13.9128(2)	59.779(1)	89.808(7)	0.191(3)	384.82(2)	64.13(7)
200	5.6484(4)	13.9089(5)	59.767(6)	89.798(7)	0.201(3)	384.29(9)	64.05(1)
150	5.6460(3)	13.9023(6)	59.769(6)	89.800(5)	0.199(5)	383.78(9)	63.96(5)
100	5.6432(7)	13.8950(3)	59.771(3)	89.802(1)	0.198(1)	383.21(3)	63.86(9)
50	5.6411(8)	13.8852(9)	59.785(8)	89.814(5)	0.185(5)	382.65(9)	63.77(7)

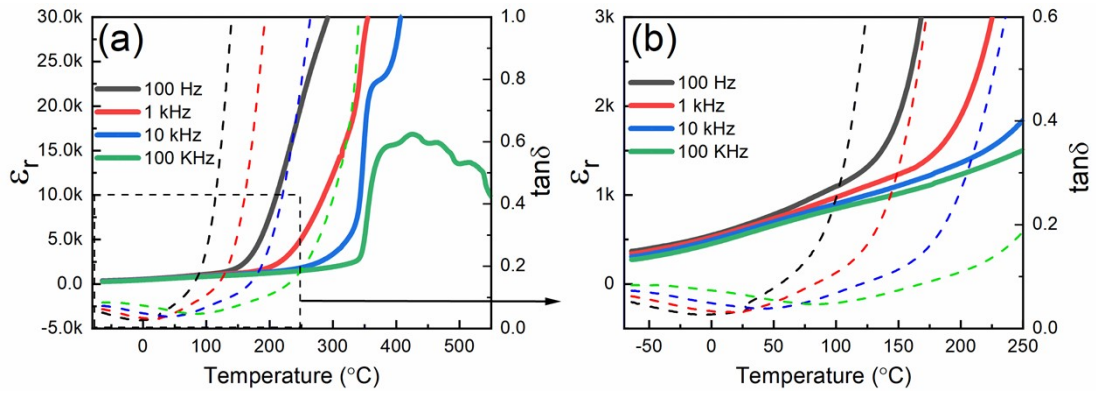


Figure S2. (a) Temperature dependence of relative permittivity (ϵ_r) and loss ($\tan \delta$) for Ar annealed pure 0.7BF-0.3BT ceramics and (b) details of relative permittivity and dielectric loss over the temperature range from -60 to 250 °C.

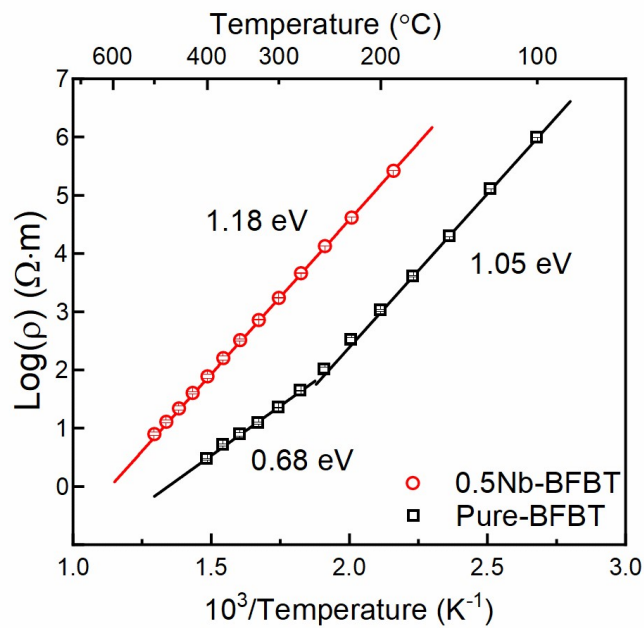


Figure S3. Arrhenius plot of resistivity of Nb-doped 0.7BF-0.3BT and Ar annealed pure 0.7BF-0.3BT ceramics.

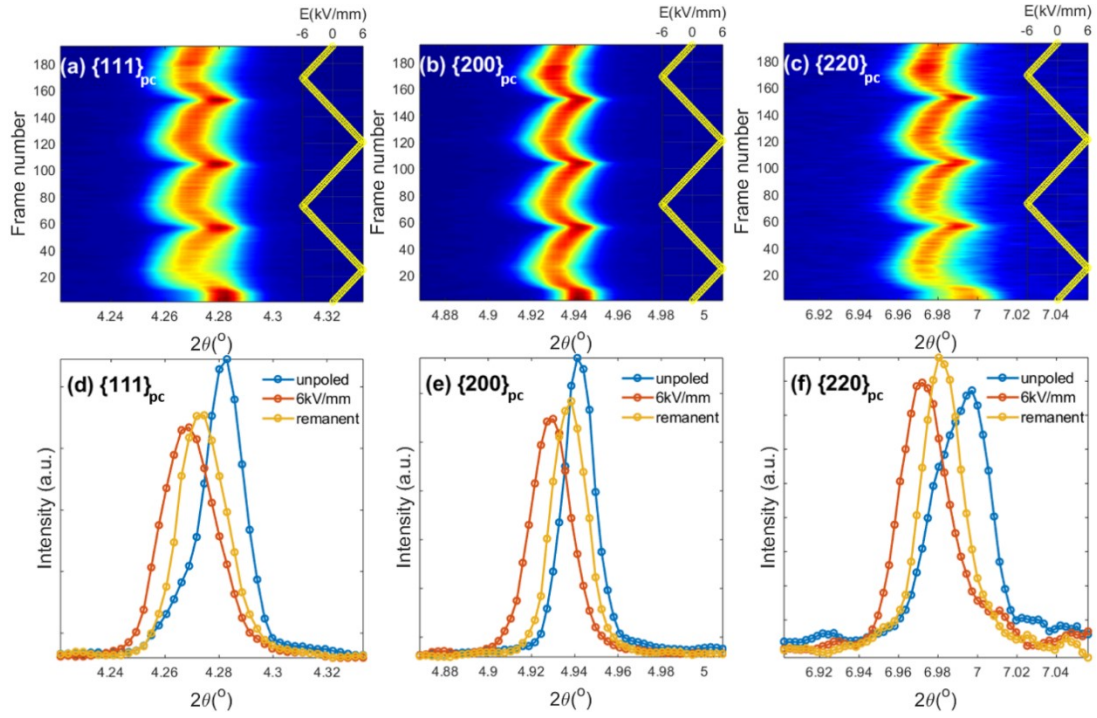


Figure S4. XRD contour plots of Nb-doped 0.7BF-0.3BT ceramics at 25 °C for the $\{111\}_{pc}$, $\{200\}_{pc}$ and $\{220\}_{pc}$ reflections under 2 bipolar electric field cycles in (a), (b) and (c) respectively, and comparison of peak profiles for the unpoled state, under an electric field of 6 kV mm⁻¹ and in the remanent state in (d), (e) and (f).

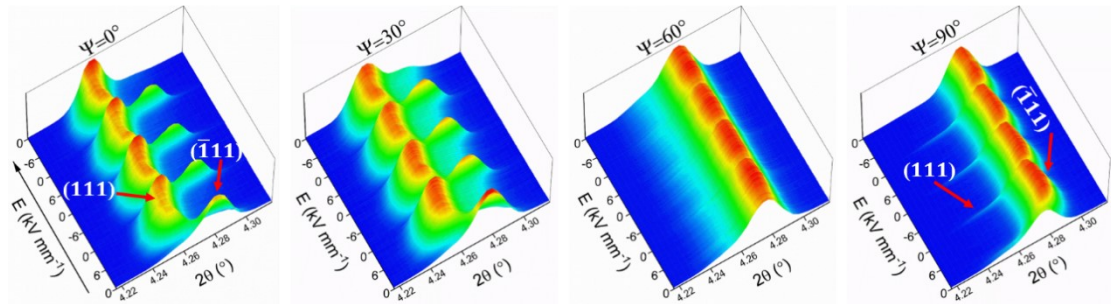


Figure S5. In-situ synchrotron XRD contour plots of the $\{111\}_{pc}$ reflections for Nb-doped 0.7BF-0.3BT ceramics at 125 °C under 2 bipolar electric field cycles, for azimuthal angles, $\psi = 0^\circ, 30^\circ, 60^\circ$ and 90° .

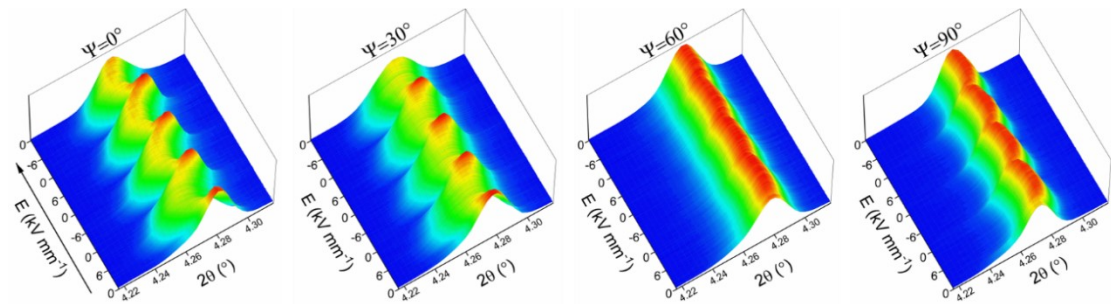


Figure S6. In-situ synchrotron XRD contour plots of the $\{111\}_{pc}$ reflections for Nb-doped 0.7BF-0.3BT ceramics at 25 °C under 2 bipolar electric field cycles, for azimuthal angles, $\psi = 0^\circ, 30^\circ, 60^\circ$ and 90° .