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Supporting Information

Advances in modification of contradictory relationship: Simultaneously realizing large piezoelectricity and high Curie temperature in potassium sodium niobate based ferroelectrics

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Table S1: Crystal structure parameters of KNNS*x*-BNZ-BF ceramics: *x*=0 and 0.03.

Parameters	***x*=0**		***x*=0.03**	
Symmetry	R Phase	T Phase	R Phase	T Phase
proportion	68.53%	31.47%	28.26%	71.74%
Space group	*R*3*m*	*P*4*mm*	*R*3*m*	*P*4*mm*
a(Å)	3.9678	3.9439	3.9572	3.9613
b(Å)	3.9678	3.9439	3.9572	3.9613
c(Å)	3.9678	4.0027	3.9572	3.9987
Alpha(°)	89.9815	90	89.9533	90

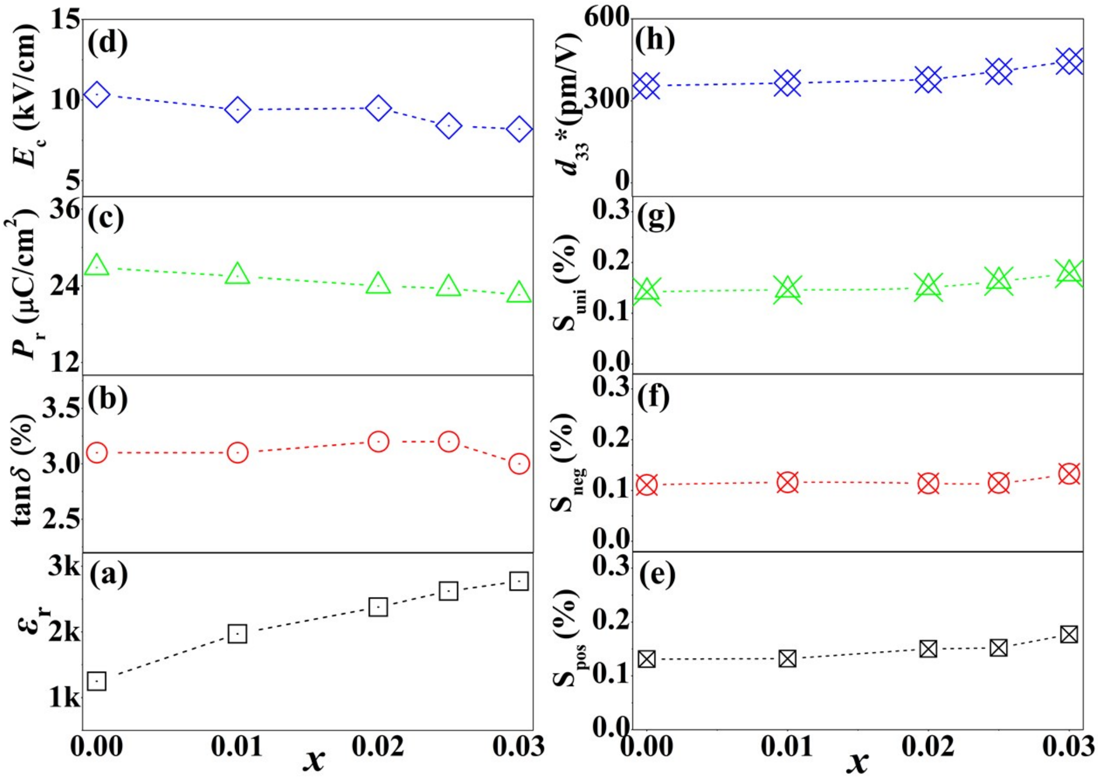

Fig.S1: (a)-(b) dielectric (ε_r, tan δ), (c)-(d) ferroelectric (P_r, E_c), (e)-(g) strain (S_{pos}, S_{neg}, S_{uni}), and (h) d_{33}* properties of KNNS$_x$-BNZ-BF ceramics.

Fig.S1(a)-(b) shows dielectric (ε_r, tan δ) properties of KNNS$_x$-BNZ-BF ceramics. The ε_r almost linearly increases from 1249 to 2770 with increasing Sb^{5+}, which benefits from the $T_{R\text{-}T}$ closing to the room temperature by adding Sb^{5+}. The tan δ of all ceramics is around 3%~3.2%, and a lower tan δ (~3%) is obtained in the ceramics with x=0.03. The ferroelectric (P_r, E_c) properties of KNNS$_x$-BNZ-BF ceramics is shown in Fig.S1(c)-(d), and the corresponding parameters are extracted from the *P*-*E* loops [See Fig. S2]. Recent publications reported that KNN-based ceramics with doping Sb^{5+} will degrade the ferroelectric properties, which lead to a slow degradation of P_r as a function of

Sb^{5+}.[1-3] The E_c presents a slight softening change with increasing Sb^{5+}, and a lower E_c is obtained in the ceramics with x=0.03. As we know, a low

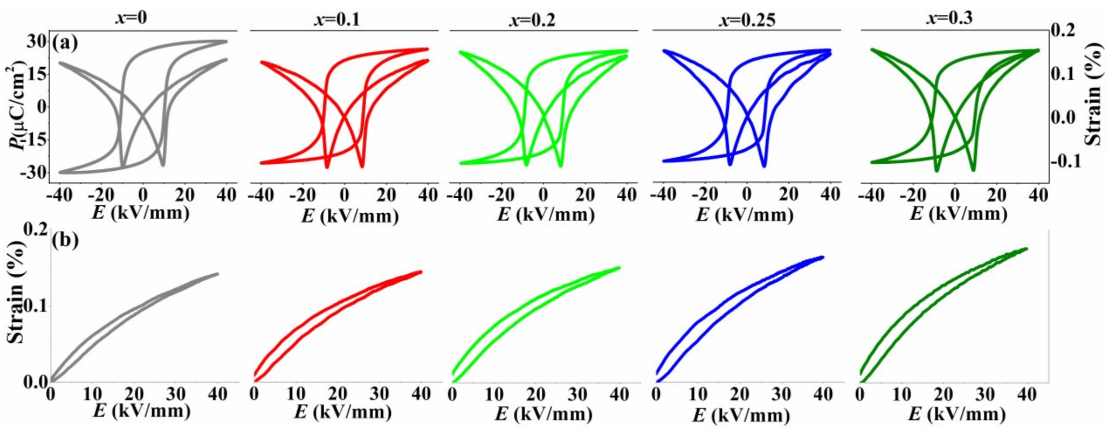

Fig.S2: (a) Ferroelectric loops, (b)Bipolar strain loops, and (c) Unipolar strain curves of KNNS$_x$-BNZ-BF ceramics.

energy barrier usually exists in the ceramics with multiphase coexistence, benefiting to making domain switching and polarization rotation easier.[4, 5] The decreasing E_c can be ascribed to the multiphase structure closing to the room temperature. The strain (S_{pos}, S_{neg}, S_{uni}) properties as a function of Sb^{5+} are plotted in Fig.S1(e)-(g), measured at 1Hz and 40 kV/cm, the S_{pos} and S_{neg} are extracted from *S*-*E* loops and S_{uni} is derived from unipolar strain curves[See Fig. S2]. S_{pos} monotonely increases (S_{pos}: 0.131%→0.177%) with adding Sb^{5+} , gaining a higher strain (S_{pos} ~0.177%) value at R-T phase zone near the room temperature [See Fig. S1(e)]. S_{neg} (S_{neg}: 0.111%→0.133%) have the similar variation with increasing Sb^{5+} [See Fig. S1(f)], indicating that non-180° domains (*eg*.,71°, 109° and 90°) increase in the R-T phase boundary.[6] As for unipolar strain

(S_{uni}:0.142%-0.178%), has a similar trend to S_{pos} as the increase of Sb^{5+}[See Fig. S1(g)], which can be explained by same contributions (eg., intrinsic piezoelectric response strain, extrinsic domain switching strain) of the S_{pos} and S_{uni}. d_{33}^* (S_{uni}/E_{max})is plotted in Fig.S1(g), the change trend highly matched the S_{uni}, that is, the d_{33}^* increases from 355 pm/V to 445pm/V as a function of Sb^{5+}. Considering the relatively high S_{neg}, S_{uni} and S_{pos} in the R-T phase boundary, the strain properties mainly originate from intrinsic and extrinsic contribution, such as the piezoelectric response strain, domain switching strain.

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