

Supporting Information

**Niobate Based Lead-Free Piezoceramics: Diffused Phase Transition
Boundary Leading to Temperature-Insensitive High Piezoelectric Voltage
Coefficient**

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I. Phase structure

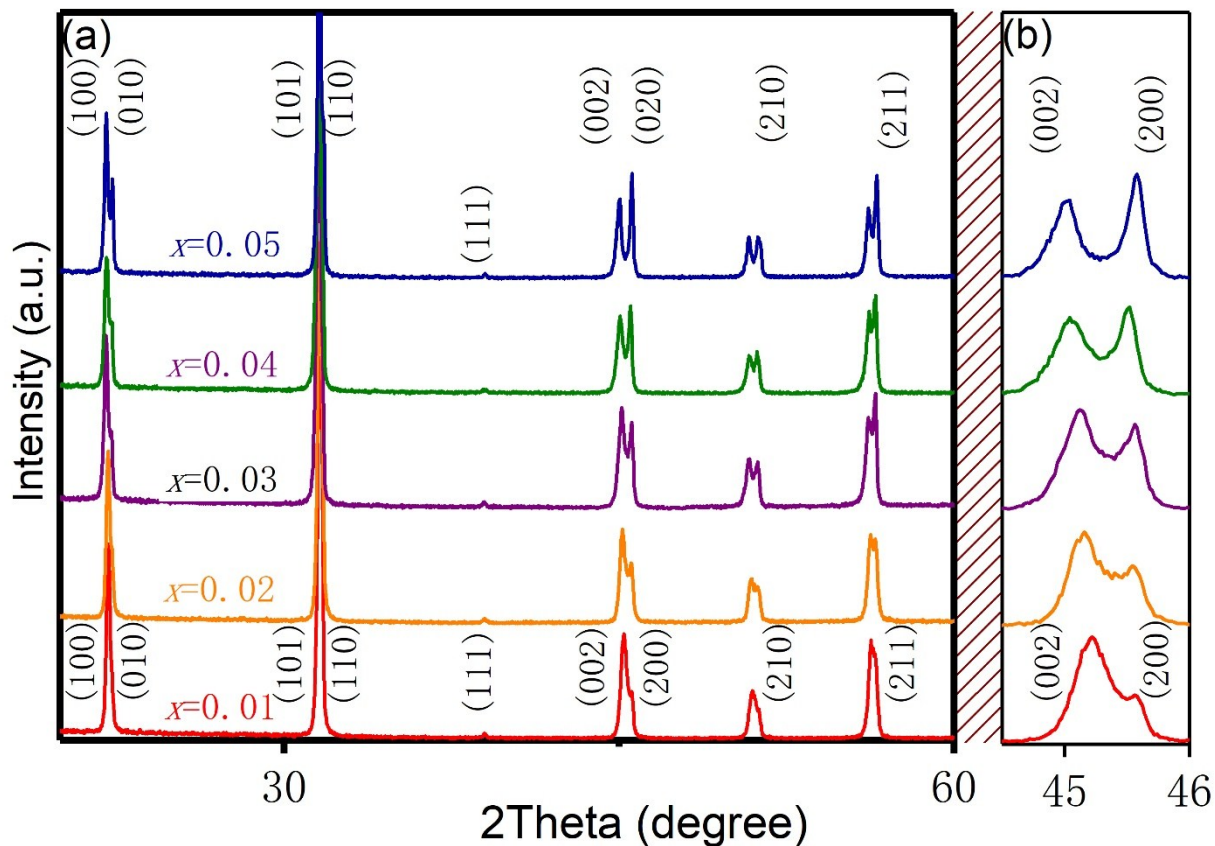


Fig. S1 Room temperature XRD patterns of the $L_x\text{KNN-6.5BZ-1BNT}$ samples

II. Mechanical property of $L_{0.02}\text{KNN-6.5BZ-1BNT}$ sample

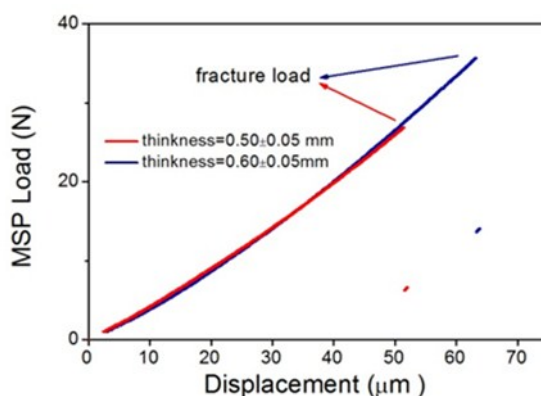


Fig. S2 MSP load-displacement curves for $L_{0.02}\text{KNN-6.5BZ-1BNT}$ sample.

It makes sense if the lead-free piezoelectric ceramics exhibit both considerable piezoelectricity and good mechanical property. And it is considered according to the experimental data as listed in Table 1 that the high density of the $L_x\text{KNN-6.5BZ-1BNT}$

favors the high k_p value. The mechanical property is considered to be highly related to the microstructure. A modified small punch (MSP) method was used to evaluate the mechanical strength according to the following **Equation S1**:

$$\sigma_f = \frac{3P}{2\pi t^2} \left[1 - \frac{1-\nu^2}{4} \times \frac{b^2}{a^2} + (1+\nu) \times \ln \frac{a}{b} \right] \quad (S1)$$

where P is the fracture load, t is the thickness of the sample, ν is the Poisson ratio, 2a is the diameter of the load-supporting hole of the lower die, and 2b is the diameter of the cylinder-shaped pressure head. The samples exhibit brittle fracture behavior. The fracture load P could be identified as an abrupt load decline occurs, before which a linear dependence of displacement is observed for the MSP load. The MSP fracture loads are $P_1=26.8$ N with $t_1=0.50$ mm, $P_2=35.6$ N with $t_2=0.60$ mm. The two observed MSP σ_f values are expected to be similar and are calculated to be 81MPa and 78MPa, respectively, which are larger than the previous reported KNN-based ceramics.^[1]

III. Translucent feature of $L_{0.02}$ KNN-6.5BZ-1BNT sample

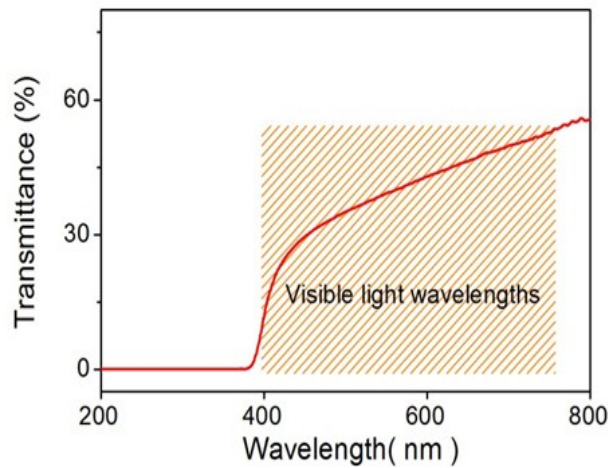


Fig. S3 Optical transmittance spectra of $L_{0.02}$ KNN-6.5BZ-1BNT sample of 0.35 mm in thickness. The optical transmittance reaches around 40% at the 633 nm wavelength.

IV. Dielectric property

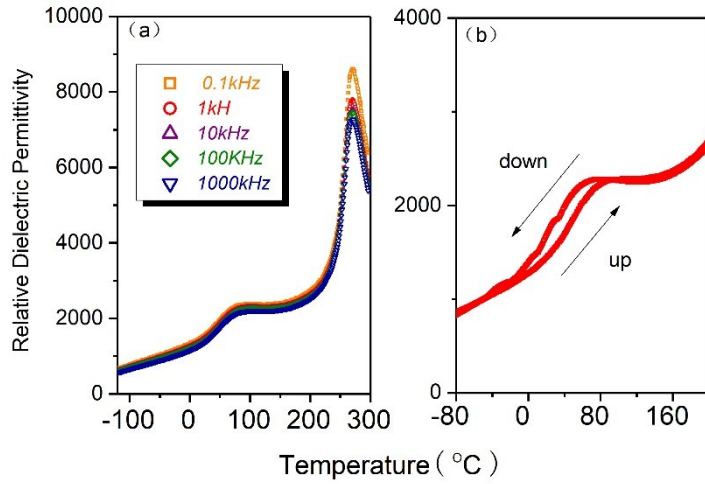


Fig. S4 (a) the temperature-dependent permittivity of the $x=0.02$ sample at different frequencies.(b) the temperature hysteresis.

V. Piezoelectric property

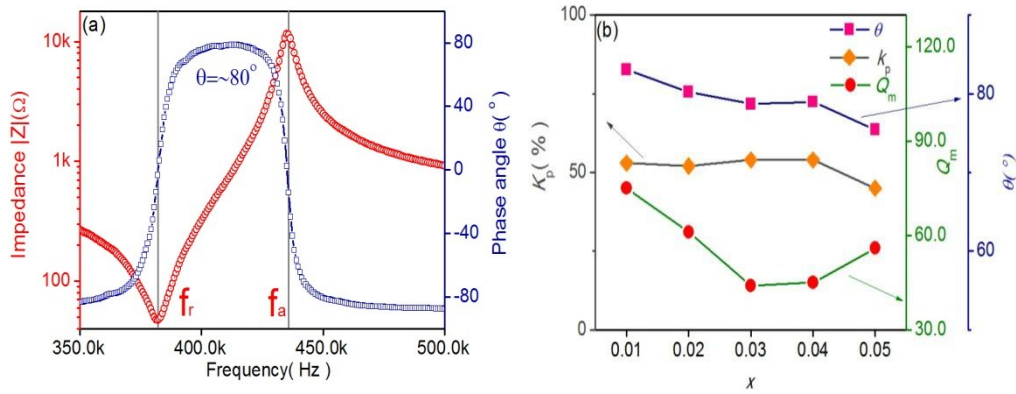


Fig. S5 (a) The frequency dependence of the impedance and phase angle of $L_{0.03}KNN-6.5BZ-1BNT$ samples. (b) Summarized results in θ , k_p and Q_m of $L_xKNN-6.5BZ-1BNT$ ceramics as functions of Li contents.

It can be seen that the sample with $x=0.03$ displays a square-like resonance peak, featured with both the resonance 381 kHz and anti-resonance frequency 435 kHz. It was calculated that the planar mode electromechanical coupling factor k_p could reach 54% according to the **Equation S2** as follows.

$$k_p = \frac{1}{\sqrt{0.395 \frac{f_r}{f_r - f_a} + 0.574}} \quad (S2)$$

VI. Comparison of temperature dependence of piezoelectricity

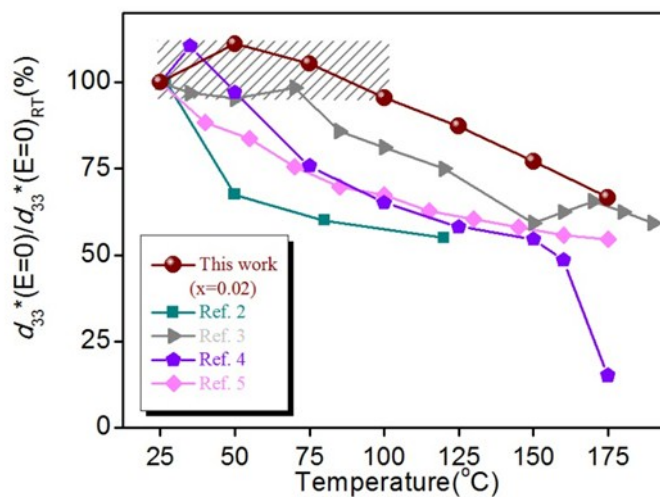


Fig. S6 Comparison of temperature dependence of small signal $d_{33}^*(E=0)$ for various ceramics as normalized to its room temperature value $d_{33}^*(E=0)_{RT}$.

VII. Temperature dependence of ferroelectric property

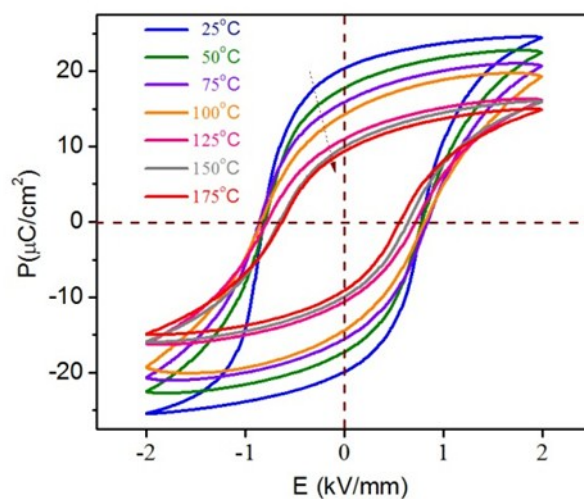


Fig. S7 P-E loops of $L_{0.02}KNN-6.5BZ-1BNT$ ceramics at different temperatures.

Reference

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