

Fig. 1S. ⁷Li solid state NMR spectra of (1-x)KNN-*x*BaLN ceramics at x = 0.005 and 0.02. Strong signal of ⁷Li around 4 ppm in both the compositions is due to the Li⁺ cation in (1-x)KNN-*x*BaLN ceramics, while the weak signal around -3 ppm (marked as $\mathbf{\nabla}$) at x = 0.02 is attributed to the presence of minor secondary phase.



Fig. 2S. XRPD patterns of (1-x)KNN-*x*BaLN ceramics (x = 0, 0.005, and 0.01) in 20 range of 44-47 degree measured at 30-450 °C after forged for 10 min at each temperature, suggesting the phase transition behavior from orthorhombic to tetragonal phase at the temperatures ranging from 185 °C to 200 °C and the presence of cubic phase above T_c .



Fig. S3. *P-E* hysteresis loops of (1-*x*)KNN-*x*BaLN ceramics with different compositions measured at 1 Hz.