Supplementary Information

Piezoelectricity of Lead-Free (K, Na)NbO3 Nanoscale Single Crystals

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S1. Material stiffness



Fig. S1 Material stiffness calculated from the force curves for KNN NRs with various compositions.

During the approach or retraction of the tip to/from the sample, the distance dependence of force when tip contacts sample, can be rendered as a datum of qualitative material stiffness. The variation trend of estimated elestic constants acquired with the same probe, *i.e.* the contact slope, is in accordance with that of resonance frequency revealed in Fig. 3(b).

S2. Domain structures



Fig. S2 Domain structures in KNN NR with K/Na ratio of 54/46 that is off MPB region, and rare domains at the nanoscale could be observed.



S3. TEM sample fabrication

Fig. S3 Graphical illustration of preparation of KNN NRs TEM samples, (a) the KNN NRs were embedded in the polymer matrix, then (b) the bulk was sliced to the flake with thickness of $\sim 10 \,\mu$ m, and (c) the TEM image of thin sprayed area.