Origin of giant piezoelectric effect in lead-free $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ single crystals

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A series of high-quality, large-sized (maximum size of $16 \times 16 \times 32$ mm$^3$) $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ ($x = 0.61$, 0.64, and 0.70 and corresponding $y = 0.58$, 0.60, and 0.63) single crystals were grown using the top-seed solution growth method. The segregation of the crystals, which allowed for precise control of the individual components of the crystals during growth, was investigated. The obtained crystals exhibited excellent properties without being annealed, including a low dielectric loss (0.006), a saturated polarization vs. electric field hysteresis loop, a giant piezoelectric coefficient $d_{33}$ ($d_{33} = 416$ pC/N, determined by the resonance method and $d_{33}^* = 480$ pC/N, measured using a piezo-$d_{33}$ meter), and a large electromechanical coupling factor, $k_{33}$ ($k_{33} = 83.6\%$), which was comparable to that of lead zirconate titanate. The reason the piezoelectric coefficient $d_{33}$ of $K_{0.39}Na_{0.61}Ta_{0.6}Nb_{0.4}O_3$ was larger than that of the other two crystals grown was elucidated through first-principles calculations. The obtained results indicated that $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ crystals can be used as a high-quality, lead-free piezoelectric material.

Piezoelectric materials are used widely in a variety of applications, including in ultrasonic transducers and piezoelectric transformers and sensor. For the last 60 years, lead zirconate titanate (PZT) ceramics were the most popular piezoelectric materials, owing to their excellent piezoelectric properties1–3. However, because lead causes significant harm to the environment and human health, efforts have been made to develop lead-free piezoelectric materials4–5. It is widely believed that potassium sodium niobate (K1−xNa0.513Li0.01−0.04)(Nb0.667Ta0.333)O3 (KNN)-based materials have great potential as replacements for lead-based piezoelectric materials because of their excellent properties and because their Curie temperatures ($T_c$) are higher than those of other lead-free materials such as barium titanate (BT) and bismuth sodium titanate (BNT)6–7.

In 2004, Y. Saito et al. reported the large piezoelectric effect in Li, Ta, Sb co-doped KNN ceramics. A major breakthrough was the fact that the piezoelectric coefficient $d_{33}$ of these KNN-based ceramics was as high as 416 pC/N8. Since then, intensive research efforts have been made to improve the piezoelectric properties of KNN-based ceramics9–16. As is known, the properties of single crystals are better than those of ceramics of the same composition. Furthermore, in order to investigate the mechanism responsible for the large piezoelectric effect observed in doped KNN and to improve the performance of KNN-based materials, it is important to grow KNN-based single crystals. Recently, several methods for growing KNN-based crystals have been reported, and each method has its advantages and disadvantages. The most widely used methods for growing KNN-based single crystals are the flux, Bridgman, and top-seed solution growth (TSSG) methods. The flux method yields crystals of good quality; however, the as-grown crystals are difficult to study, owing to their small size17–19. Chen et al. successfully grew Li:KNN single crystals with a large $d_{33}$ ($405$ pC/N) using the Bridgman method; however, the single crystals contained oxygen vacancies and defects, which led to a high leakage current, as was evident from their polarization vs. electric field hysteresis loops20. Zheng et al. grew Li, Ta, Sb-doped KNN single crystals using the TSSG method and investigated their piezoelectric properties. They were able to obtain $K_{0.51}Na_{0.49}Nb_{0.74}Ta_{0.26}O_3$ ($d_{33} = 200$ pC/N, $k_{33} = 82.7\%$), $(K_{0.65}Na_{0.35}Li_{0.01~0.04})(Nb_{0.667}Ta_{0.333})O_3$ ($d_{33} = 354$ pC/N, $k_{33} = 81.5\%$), and $(K_{0.51}Na_{0.49}Li_{0.02~0.03})(Nb_{0.7}Ta_{0.3}Sb_{0.3})O_3$ ($d_{33} = 172$ pC/N, $k_{33} = 52.3\%$)21–23. For the lack of continuous changes in the composition and doping ions, the research of the contribution of doping ions on the piezoelectric 

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properties of KNN is difficult. Furthermore, there have been few microscopic studies on the physical mechanism responsible for the improvement in the piezoelectric properties after doping with Ta.

In this paper, we report the successful growth of a series of high-quality, large-sized $\text{K}_{1-x}\text{Na}_x\text{Ta}_{1-y}\text{Nb}_y\text{O}_3$ (KNTN) single crystals using the TSSG method. The segregation of the crystals, which allowed for precise control over the individual components of the crystals, was investigated. The as-grown crystals exhibited excellent properties, including a saturated hysteresis loop, a giant piezoelectric coefficient, $d_{33}^*$ (480 pC/N), and a large electromechanical coupling factor, $k_{33}$ (83.6%); the values of these parameters were comparable to those of PZT. Finally, the origin of the high piezoelectric effect observed in the crystals was elucidated through first-principles calculations.

Results and Discussion

Photographs of the as-grown KNTN single crystals are shown in Fig. 1. The dimensions of the largest KNTN crystal grown were $16 \times 16 \times 32 \text{ mm}^3$, which are much greater than those of previously reported KNTN crystals. The crystals were transparent at temperatures higher than their Curie temperature ($T_C$), and no cracks were observed in them. At room temperature, the crystals were milky white, because of the presence of polydomains, which scatter light. The KNTN crystals were shaped like a square with round corners, and the surfaces of the crystals were (100)$_c$ and (010)$_c$ faces.

The segregation of the crystals allowed for precise control of the individual components of the crystals during the growth process. The relationships between the composition of the KNTN crystals; the ratio of the potassium and sodium concentrations in the melt; the ratio of the tantalum and niobium concentrations in the melt; the segregation coefficient of potassium, sodium, tantalum, and niobium; and the growth temperature ($T_g$) are shown in Table 1. The segregation coefficient of potassium ($S^K$) was calculated from the equation: $S^K = C^K/C_{m}^{K}$, where $C^K$ is K concentration in single crystal; $C_{m}^{K}$ is K concentration in melt. The similar calculations were performed for $S^{Na}$, $S^{Ta}$, $S^{Nb}$. The segregation of ion was determined by their properties. Moreover, it was also determined by ion concentration in melt and the condition of growth. With an increase in the tantalum content in the melt, the

![Figure 1. Photographs of the as-grown KNTN single crystals.](image)

Table 1. Segregation of potassium, sodium, tantalum, and niobium and the growth temperatures of $\text{K}_{1-x}\text{Na}_x\text{Ta}_{1-y}\text{Nb}_y\text{O}_3$ crystals.
The growth temperature increased from 1193 °C to 1277.5 °C. On the other hand, the segregation coefficient of tantalum decreased from 3.700 to 2.333, which in accordance with the phase diagram of the KTaO3-KNbO3 system. Further, the segregation coefficient of potassium increased from 0.400 to 0.520, while that of sodium decreased from 2.800 to 2.560. This was in contradiction to the phase diagram of the KNbO3-NaNbO3 system and mean that the doping of Ta into the KNN system affected the segregation of K and Na.

The XRD patterns indicated that the crystals had pure perovskite-like structures and did not contain any secondary phases (Fig. 2). The as-grown KNTN single crystals were in the orthorhombic phase at room temperature. From the XRD data, the lattice parameters of the KNTN crystals were determined using the software Jade 6.0; the results are shown in Table 2. The (202) and (020) peaks moved to the high degrees decreases in x and y, resulting in increases in the lattice parameters a and c. These changes in the lattice parameters of the KNTN crystals indicated that the volume of the lattice cell increased with the decrease in x and y. The cation radius of the tantalum (Ta5+) is slightly smaller than that of the niobium (Nb5+), while the volume of the as-grown crystals increase with decrease of Nb concentration (as Table 2 shown). Thus the main reason for the increase in the volume was the fact that the cation radius of potassium (K+) is larger than that of sodium (Na+).

The temperature dependence of the dielectric constant of the K1−xNa0.35Nb0.65O3 crystals at 1 kHz is shown in Fig. 3. The two peaks in the curves represent the orthorhombic-tetragonal phase-transition temperature (Tc) and the Curie temperature (Tc), respectively; the values of these temperatures were consistent with the XRD results and suggested that the crystals were in the orthorhombic phase at room temperature. As can be seen from the data listed in Table 3, with the tantalum and sodium content varying, the value of Tc decreased from 84 °C to 39 °C. Concomitantly, the value of the Curie temperature (Tc) decreased from 228 °C to 161 °C. According to the data of pure KNN single crystals, we can determine that the value of Tc of the KNN crystals slightly increased with decrease of Na concentration. However, the Tc of KTN single crystals gotten by the function Tc = 676x + 32 K (x is the Nb concentration in KTN crystals) decreased with doping Ta. Thus, the variation of Tc of KTN single crystals with the increase in the tantalum and potassium contents, and is larger than that of the pure KNN single crystals. The result is mainly reasonable because Ta could move the tantalum decreased from 3.700 to 2.333, which in accordance with the phase diagram of the KTaO3-KNbO3 system and mean that the doping of Ta into the KNN system affected the segregation of K and Na.

Further, the segregation coefficient of potassium increased from 0.400 to 0.520, while that of sodium decreased from 0.400 to 0.520, which suggests that as-grown crystals were of high quality. The values of the coercive field (Ee), remanent polarization (Pr), and spontaneous polarization (Ps) of the KNTN single crystals as well as those of the pure KNN single crystals are listed in Table 4. The Ee, Pr, and Ps values of the KNTN crystals were smaller than those of pure KNN single crystals. The result is in good agreement with the difference between Ta5+ and Nb5+ in unit...
cell, Ta$^{5+}$ and Nb$^{5+}$ ions were randomly distributed in the B-sites. Because Nb$^{5+}$ ions moving along the 12 directions of spontaneous polarization mainly contribute to the $P_r$, the $P_r$ decreased with doping Ta. The values of the room-temperature leakage current density ($J$) along the $[001]_C$ direction of the as-grown KNTN crystals are

![Figure 3. Temperature dependence of the dielectric constant of the $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ crystals at 1 kHz.](image)

| Crystal        | $T_{D-P}$ ($^\circ$C) | $T_C$ ($^\circ$C) | $\varepsilon_r$ at $T_{room}$ | $\tan\delta$ at $T_{room}$ (%) |
|----------------|-----------------------|-------------------|-------------------------------|--------------------------------|
| $K_{0.3}Na_{0.7}Ta_{0.3}Nb_{0.6}O_3$ | 84                    | 228               | 369                           | 0.7                            |
| $K_{0.36}Na_{0.64}Ta_{0.40}Nb_{0.60}O_3$ | 58                    | 209               | 609                           | 0.7                            |
| $K_{0.39}Na_{0.61}Ta_{0.42}Nb_{0.58}O_3$ | 39                    | 161               | 670                           | 0.6                            |
| $K_{0.334}Na_{0.666}NbO_3$  | 174                   | 395               | 68                            | 0.4                            |
| $K_{0.5}Na_{0.5}NbO_3$  | 192                   | 410               | 1015                          | 1                              |

Table 3. The $T_{D-P}$, $T_C$, dielectric constant ($\varepsilon_r$), and dielectric loss ($\tan\delta$) values of the KNTN single crystal at a frequency of 1 kHz.

![Figure 4. (a) $P$-$E$ hysteresis loops of the KNTN crystals at room temperature at a frequency of 200 Hz. (b) Values of the leakage current density ($J$) at room temperature along the $[001]_C$ direction of the KNTN crystals.](image)

| Crystal        | $E_c$ (kV/cm) | $P_r$ ($\mu$C/cm$^2$) | $P_s$ ($\mu$C/cm$^2$) |
|----------------|--------------|------------------------|------------------------|
| $K_{0.3}Na_{0.7}Ta_{0.3}Nb_{0.6}O_3$ | 5.52         | 6.11                   | 9.14                   |
| $K_{0.36}Na_{0.64}Ta_{0.40}Nb_{0.60}O_3$ | 5.05         | 5.37                   | 10.34                  |
| $K_{0.39}Na_{0.61}Ta_{0.42}Nb_{0.58}O_3$ | 6.73         | 6.59                   | 9.86                   |
| $K_{0.334}Na_{0.666}NbO_3$  | 11.59        | ~12                    | ~12                    |
| $K_{0.455}Na_{0.545}NbO_3$  | 11.11        | ~12                    | ~12                    |

Table 4. Coercive field ($E_c$), remanent polarization ($P_r$), and spontaneous polarization ($P_s$) values of the $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ single crystals.
shown in Fig. 4b. The leakage currents for the KNTN crystals which were not annealed at high temperature had an order of magnitude of ~10⁻⁰ A/cm², which is far lower than that for KNN crystals grown by the flux method (10⁻¹ to 10⁻⁴ A/cm²)30,31. This result suggested that there were less O²⁻ vacancies in the crystals grown by TSSG method26, which was another indication of the high quality of the KNTN single crystals grown in this study. Because of the more O²⁻ vacancies of K₀.₃₀Na₀.₇₀Ta₀.₃₇Nb₀.₆₃O₃ caused by the lower melting temperature, the f₀ of that was one order of magnitude larger than other two single crystals.

The piezoelectric vibrators were cut along the [001], [101], and [110] directions, and their impedance spectra (magnitude of impedance |Z| and phase angle, θ, vs. frequency) were measured, so that their piezoelectric parameters could be calculated. A typical longitudinal extension response for the k₃₃ resonator of K₀.₃₉Na₀.₆₁Ta₀.₄₂Nb₀.₅₈O₃ single crystal was shown in Fig. 5. For this k₃₃ resonator, a resonance frequency (f₀) was 631 kHz while the antiresonance frequency (fₐ) was 1.103 MHz. The values of the elastic compliance constant s₃₃, electromechanical coupling factor k₃₃, and piezoelectric coefficient d₃₃ of the samples were calculated by substituting the measured long-size l, f₀ and fₐ into the following equation: k₃₃² = (π fₐ/(2f₀)) tan[π (fₐ − f₀)/(2f₀)], s₃₃ = 1/(ρ(2f₀²)), s₃₃ = s₃₃/[1 − k₃₃²], d₃₃ = k₃₃ √(ρ s₃₃). The maximum phase angle was approximately 75°; this was indicative of the existence of a polydomain state (domain engineered). The similar measurement was performed for other k₃₃, k₃₁, and k₄₄ resonators. The k₃₃ values of the samples were between 76.4% and 83.6% and larger than those reported previously for KNN-TL and PZT5A. Further, the giant piezoelectric coefficient d₃₃ of the K₀.₃₉Na₀.₆₁Ta₀.₄₂Nb₀.₅₈O₃ crystal was 416 pC/N, as determined by the resonance method, while d₃₃ was 480 pC/N, as measured with a piezo-d₃₃ meter; these values are larger than those for other two as-grown single crystals and KNN-TL. As can be seen from the data, the values of most of the parameters of K₀.₃₉Na₀.₆₁Ta₀.₄₂Nb₀.₅₈O₃ which were greatly influenced by temperature were comparable to those of PZT5A.

As shown in Table 5, the piezoelectric coefficient d₃₃ of the K₀.₃₉Na₀.₆₁Ta₀.₄₂Nb₀.₅₈O₃ crystal was larger than those of the K₀.₃₇Na₀.₇₀Ta₀.₃₇Nb₀.₆₃O₃ and K₀.₃₆Na₀.₆₄Ta₀.₄₀Nb₀.₆₀O₃ crystals. This can be attributed to the polymorphic phase transition (PPT) which was lowered to room temperature by Ta, when the K/Na ratio was far away from 0.₅/0.₅32,33. However, the microscopic physical mechanism responsible was not involved. To elucidate the origin of the high piezoelectric effect in the microcosmic, we used first-principles calculations to calculate the free energy of the lattice cell.

Spontaneous polarization occurs in twelve directions along [001], in the orthorhombic (mm2)-phase single crystals, while it occurs in six directions along [001], for the tetragonal (4 mm) -phase single crystals (shown in Fig. 6). When an electric field was applied along the z-axis, the directions in which spontaneous polarization occurred in the orthorhombic phase changed to those corresponding to the tetragonal phase. In addition, according to a previous study, the reason the piezoelectric coefficient d₃₃ of the grown crystals was large was that the polarization directions were rotated and not stretched34. The internal energies of the K₁₋ₓNaₓTa₁₋ₓNbₓO₃ crystals corresponding to the different phases (orthorhombic phase, Uₒ, and tetragonal phase, Uₜ) were calculated through first-principles calculations. For this, we calculated the internal energies of potassium tantalate (KTO₃, KT), potassium niobate (KNO₃, KN), sodium tantalate (NaTO₃, NT), and sodium niobate (NaNBO₃, NN), as well as the weighted averages of the individual coefficients. The obtained results are shown in Table 6.

It can be seen that ΔU decreased from 3.0241 eV Å³ to 2.5666 eV Å³ with an increase in the Ta fraction from 0.37 to 0.42 and an increase in the K fraction from 0.30 to 0.39. Further, the difference in the free energies of the orthorhombic and tetragonal phases of K₀.₃₉Na₀.₆₁Ta₀.₄₂Nb₀.₅₈O₃ in the absence of an electric field was the lowest. Thus, when an electric field was applied, the domain of K₀.₃₉Na₀.₆₁Ta₀.₄₂Nb₀.₅₈O₃ which was at PPT temperature, was the easiest to rotate, which can enhance piezoelectric properties34. The conclusion was the same as that calculated by Landau – Devonshire model in BaTiO₃ – based materials37,38 and PZT-based materials39. As a result, its piezoelectric coefficient was larger than those of K₀.₃₀Na₀.₇₀Ta₀.₃₇Nb₀.₆₃O₃ and K₀.₃₆Na₀.₆₄Ta₀.₄₀Nb₀.₆₀O₃.

Conclusions

In this study, a series of large-sized (size of largest crystal = 16 × 16 × 32 mm³) orthorhombic K₁₋ₓNaₓTa₁₋ₓNbₓO₃ (x = 0.₆₁, 0.₆₄, and 0.₇₀ and corresponding y = 0.₅₈, 0.₆₀, and 0.₆₃) single crystals were grown using the TSSG method. The crystals exhibited excellent dielectric, piezoelectric, and ferroelectric properties, including

Figure 5. Room-temperature k₃₃ resonator impedance spectra of K₀.₃₉Na₀.₆₁Ta₀.₄₂Nb₀.₅₈O₃ single crystal.
A giant $d_{33}^\ast$ (480 pC/N) and a large $k_{33}$ (83.6%). The leakage currents of the $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ crystals were very low and of the order of $10^{-5}$ A/cm². The piezoelectric properties of the $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ crystals improved with an increase in the potassium and tantalum contents when the phase of the $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ crystals was orthorhombic. Finally, the difference in the free energies of the orthorhombic and tetragonal phases of $K_{0.39}Na_{0.61}Ta_{0.42}Nb_{0.58}O_3$ in the absence of an electric field was $2.5666$ eV/Å³; this was determined by first-principles calculations. This crystal was the smallest of the grown single crystals. Further, its domain was the easiest to rotate. Therefore, it exhibited the best piezoelectric properties.

### Table 5. Piezoelectric parameters of the as-grown KNTN single crystals poled along $[001]$, and having tetragonal $4mm$ symmetry.

| Piezoelectric materials | $s_{E1}$ (10⁻¹² m²/N) | $s_{E33}$ (10⁻¹² m²/N) | $s_{D11}$ (10⁻¹² m²/N) | $s_{D33}$ (10⁻¹² m²/N) |
|-------------------------|-------------------------|-------------------------|-------------------------|-------------------------|
| $K_{0.30}Na_{0.70}Ta_{0.37}Nb_{0.63}O_3$ | 10.9 | 14.8 | 9.17 | 6.14 |
| $K_{0.36}Na_{0.64}Ta_{0.40}Nb_{0.60}O_3$ | 12.7 | 19.2 | 9.98 | 6.60 |
| $K_{0.39}Na_{0.61}Ta_{0.42}Nb_{0.58}O_3$ | 14.5 | 26.6 | 11.01 | 8.01 |
| KNN-TL²² | 17.2 | 27.0 | 13.4 | 9.08 |
| PZT5A²² | 16.4 | 18.8 | 14.4 | 9.46 |

### Table 6. Internal energies of the $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ crystals.

| Piezoelectric materials | $U_o$ (eV/Å³) | $U_t$ (eV/Å³) | $\Delta U = U_t - U_o$ (eV/Å³) |
|-------------------------|--------------|--------------|-------------------------------|
| $K_{0.30}Na_{0.70}Ta_{0.37}Nb_{0.63}O_3$ | 50.7596 | 53.7836 | 3.0241 |
| $K_{0.36}Na_{0.64}Ta_{0.40}Nb_{0.60}O_3$ | 49.6940 | 52.4140 | 2.7200 |
| $K_{0.39}Na_{0.61}Ta_{0.42}Nb_{0.58}O_3$ | 49.0279 | 51.5945 | 2.5666 |

Figure 6. Directions of spontaneous polarization of the KNTN crystals in the orthorhombic and tetragonal phases: (a) spontaneous polarization directions of the orthorhombic phase and (b) spontaneous polarization directions of the tetragonal phase.

Table 6. Internal energies of the $K_{1-x}Na_xTa_{1-y}Nb_yO_3$ crystals.
Methods

As mentioned above, the KNTN single crystals were grown by the TSSG method. The raw materials used were powders of K₂CO₃ (99.99%), Na₂CO₃ (99.99%), Ta₂O₅ (99.99%), and Nb₂O₅ (99.99%). They were weighted to obtain a composition of K₁−xNaₓTa₂Oₙ−xNb₂O₅ with 10 mol% excess of K₂CO₃ and Na₂CO₃ ((Kₓ−x)(Ta₂O₅ + Nb₂O₅) = 1:1:1) as the self-flux. The raw materials were mixed with ethanol, ball-milled for 24 h, and subsequently dried in an oven at 85 °C to volatilize the ethanol. Then, the mixture was calcined at 950 °C for 6 h to synthesize a KNTN polycrystal. The polycrystal was melted in a medium-frequency induction furnace at 1250 °C ~ 1375 °C, which is ~100 °C higher than the temperature for crystal growth, in order to eliminate the residual carbon dioxide and mix the compounds at the atomic level. Then, the temperature was decreased to the growth temperature, and a single crystal began to grow on a [001]c seed that was cut from a high-quality potassium tantalate niobate crystal. During crystal growth, the rotational and pulling rates were 15 r/min and 0.25 mm/h, respectively. After the completion of the growth process, the as-grown crystal was cooled to the room temperature at 35 °C/h.

The compositions of the as-grown crystals were determined by electron microprobe analysis (EPMA-1720, Shimadzu, Kyoto, Japan). The structures of the crystals were confirmed by X-ray diffraction (XRD) analyses (XRD-6000, Shimadzu, Kyoto, Japan). The crystals were oriented using a Laue X-ray machine. The [001]c in pseudo cubic structure structure surfaces of the samples were covered with silver electrodes, and the dielectric properties of the samples were measured as functions of the temperature using an inducance — capacitance — resistance (LCR) meter (E4980A, Agilent Technologies, Santa Rosa, CA). The polarization vs. electric field (P−E) hysteresis loops of the crystals were measured at 200 Hz using the ferroelectric test system (Precision Premier II, Radiant Technology, Inc., Albuquerque, NM, USA); the leakage current densities of the crystals were recorded using the same measurement instrument. Cuts of the crystals with dimensions similar to those mentioned in IEEE standards were poled in silico oil at a temperature of T₀ = 10 °C under an electric field of 30 kV/cm. The resonance and antiresonance frequencies were measured using an HP 4294 A impedance phase analyzer. The piezoelectric coefficients and electromechanical coupling factors were determined at the resonance and antiresonance frequencies, according to the IEEE standards. The piezoelectric constant d₃₃* was measured using a piezo-d₃₃ meter (Zj-3A, Institute of Acoustics, Academic Sinica, Beijing, China).

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Author Contributions
H.T. and Z.Z. generated the idea. H.T. and X.M. wrote the manuscript text. X.M. and C.H. designed the experimental schemes and grew the crystals. X.M. determined the piezoelectric and dielectric properties. P.T. determined the free energies by first-principles calculations. X.C. and G.S. confirmed the XRD spectrum. R.Z. measured the polarization vs. electric field (P-E) hysteresis loops. All authors discussed the results and substantially contributed to the manuscript.

Additional Information
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