Supplementary Materials for

"Significant performance enhancement of Nd-doped $Pb(In_{0.5}Nb_{0.5})O_3$-$PbTiO_3$ ferroelectric crystals"

Junjie Xiong[a], Yuequn Wang[c], Xiaoming Yang[a], Wenjie Zhang[a,b], Zujian Wang[a], Rongbing Su[a], Xifa Long[a], Ying Liu*[ab], Chao He *[ab]

[a] *Key Laboratory of Optoelectronic Materials Chemistry and Physics, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, 350002, China.*

[b]*Ganjiang Innovation Academy, Chinese Academy of Sciences, Ganzhou, 341000, China.*

[c]*Hangzhou Applied Acoustics Research Institute, Hangzhou, 310012, China*

**Corresponding Authors. E-mails: hechao@fjirsm.ac.cn (C. He), liuying@fjirsm.ac.cn (Y. Liu)*

Fig. S1 shows the powder XRD patterns of the PIN-0.33PT and Nd-PIN-0.33PT single crystals, showing perovskite structure. The enlarge of $(200)_c$ diffraction peak exhibit the broaden peaks (the right in Fig. S1) , indicating near MPB region.

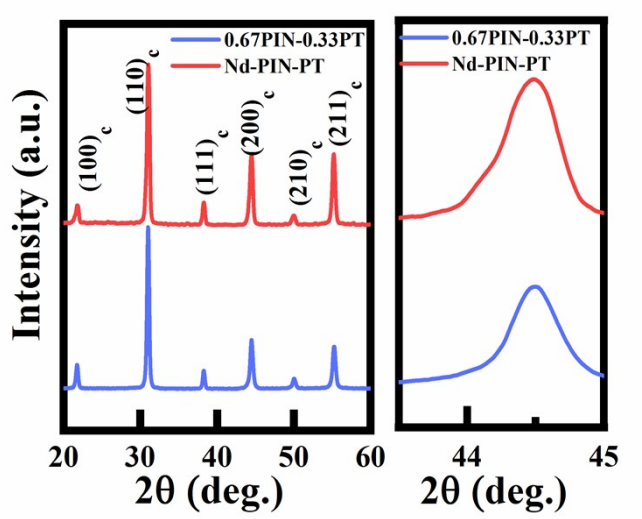

FIG. S1 The powder XRD patterns of PIN-0.33PT and Nd-PIN-0.33PT single crystals.

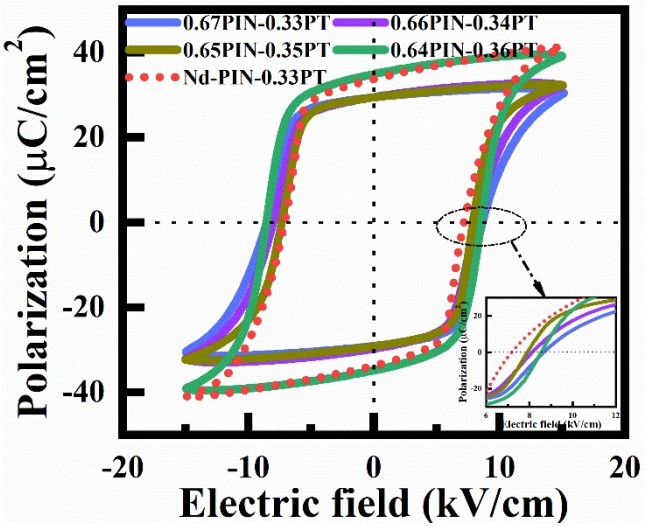

FIG. S2 The comparison of *P-E* hysteresis loops of $[001]_c$-oriented PIN-PT and Nd-PIN-0.33PT crystals at room temperature.

Fig. S2 shows the polarization-electric field (*P-E*) hysteresis loops of $[001]_c$-oriented PIN-PT and Nd-PIN-0.33PT crystals at room temperature. It can be seen that the value of coercive field (E_c) of Nd-PIN-0.33PT crystals is the smaller than PIN-PT crystals. In addition, the value of remanent polarization (P_r) of Nd-PIN-0.33PT crystals

shows high level. The low E_c and high P_r are beneficial to high piezoelectric performance.

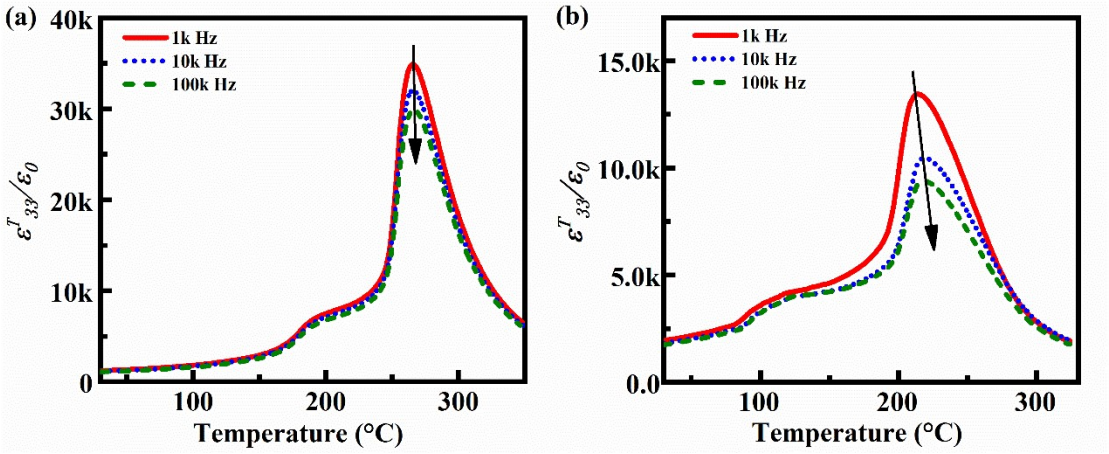

FIG. S3 Temperature dependence of $\varepsilon^T_{33}/\varepsilon_0$ of unpoled $[001]_c$-oriented (a) PIN-0.33PT and (b) Nd-PIN-0.33PT crystals.

Fig. S3 shows the temperature dependence of $\varepsilon^T_{33}/\varepsilon_0$ of unpoled $[001]_c$-oriented PIN-0.33PT and Nd-PIN-0.33PT crystals. The temperature of the maximum dielectric permittivity (T_m) shifts to higher temperature with increasing frequency, showing the feature of dielectric relaxor behavior.

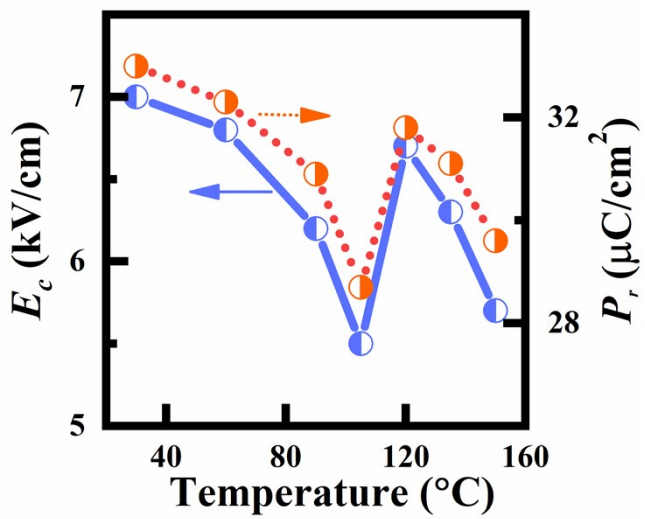

FIG. S4 Temperature dependence of E_c *and* P_r of $[001]_c$-oriented Nd-PIN-0.33PT crystals.

Fig. S4 shows the temperature dependence of E_c *and* P_r of $[001]_c$-oriented Nd-PIN-0.33PT crystals. It can be observed that the values of E_c and P_r decline first with increasing temperature from 30 °C to 105 °C, then increase significantly from 105 °C to 120 °C, finally then decrease with further increasing temperature.

Fig. S5 shows the domain structure of $[001]_c$-oriented Nd-PIN-0.33PT crystals as a function of temperature. It is interesting to note that there are no obvious changes of domain structure as the temperature increase from 30 °C to 90 °C, while the domains start to change after 120 °C, which corresponds to the depolarization temperature (T_d) for thermal stability of piezoelectric performance.

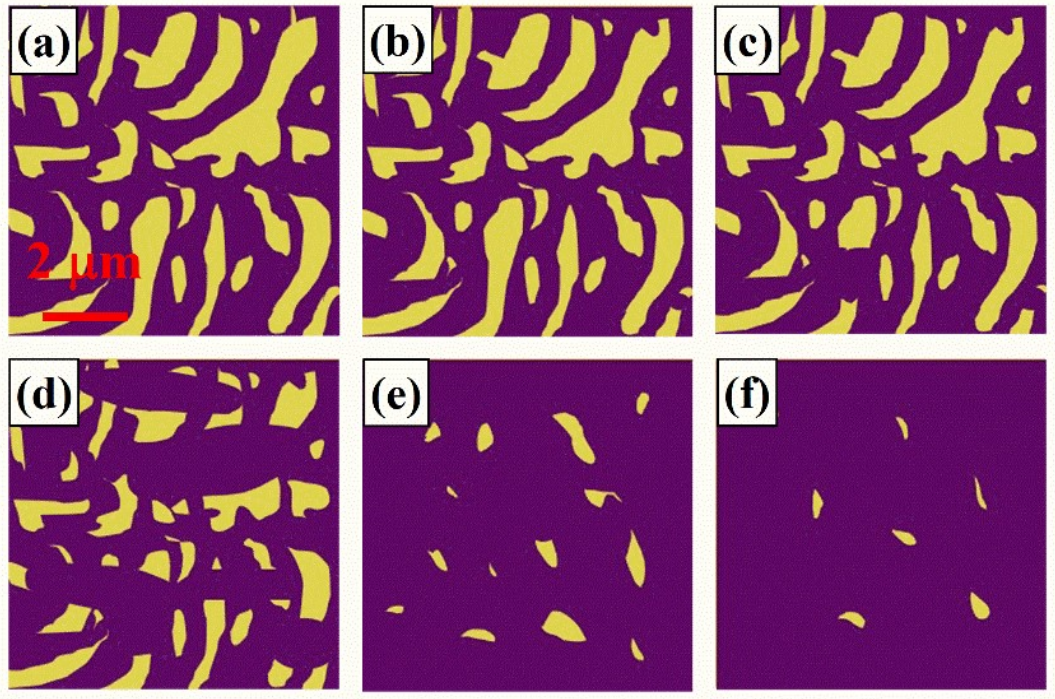

FIG. S5 The out-of-plane domain images of $[001]_c$-oriented Nd-PIN-0.33PT crystals as a function of temperature (a) 30 °C, (b) 60 °C, (c) 90 °C, (d) 120 °C, (e) 150 °C, (f) 180 °C.