

Supporting Information:

Thermal enhanced piezoelectricity via superstructure in $\text{Ca}_2\text{Nb}_2\text{O}_7$ single crystal with ultra-high Curie temperature

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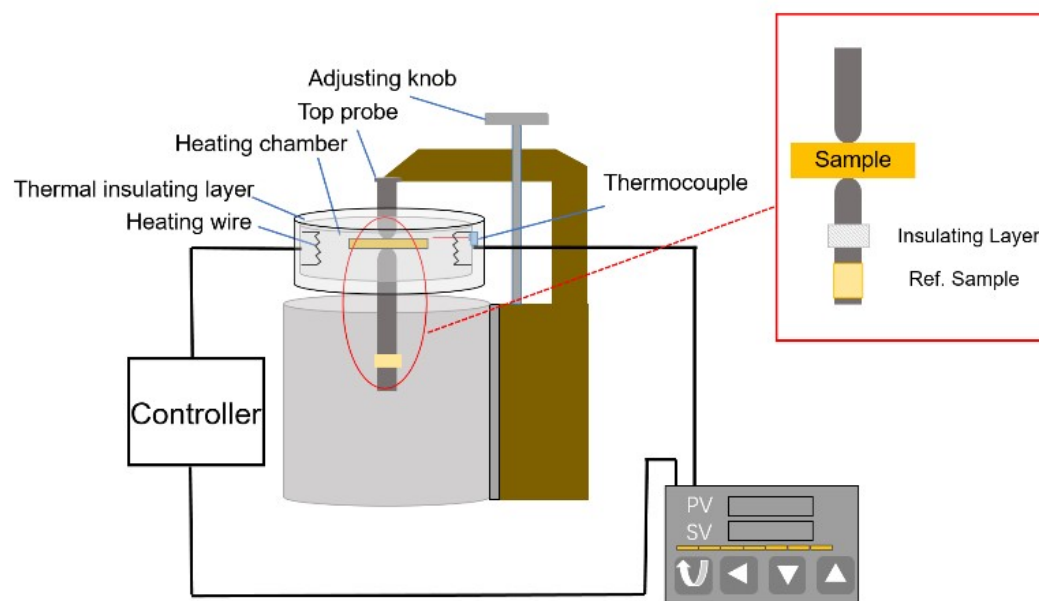


Fig. S1. Schematic illustration of the high-temperature d_{33} meter.

TEM analyzing

Fig. S2 (a) and (d) show high-resolution TEM (HRTEM) images of $\text{Ca}_2\text{Nb}_2\text{O}_7$ single crystals along the b -axis. Clear lattice fringes can be observed in these two images. The lattice fringes marked in the white square are enlarged in the Fig. S2 (a), which shows that each lattice fringe contains four layers of sub-lattice fringes. The thickness of the 4-layer of sub-lattice fringes is 1.277 nm. Fig. S2 (c) shows the crystal structure of $\text{Ca}_2\text{Nb}_2\text{O}_7$ observed along the b -axis. A 4-layer octahedron alternately repeats itself along the a -axis and adjacent two 4-layer octahedrons relatively shift $c/2$ along the c -axis, which is consistent with the four layers of sub-lattice fringes observed by HRTEM in Fig. S2 (a) and thus proves the reliability of the model of PLS with $\text{Ca}_2\text{Nb}_2\text{O}_7$ as $n=4$. The corresponding selected area electron diffraction (SAED) pattern shown in Fig. S2 (b) confirms the single-crystal structure of $\text{Ca}_2\text{Nb}_2\text{O}_7$, and the marked directions correspond to (200), (011), and (211) planes. In Fig. S2 (d), the distance between two adjacent lattice fringes is 0.359 nm, which is close to 0.352 nm of the d-spacing for the (302) plane of $\text{Ca}_2\text{Nb}_2\text{O}_7$, as shown in the Fig. S2 (c). The diffraction spot pattern in the SAED pattern shown in Fig. S2 (e) can be indexed to (302), (010), and (312) planes.

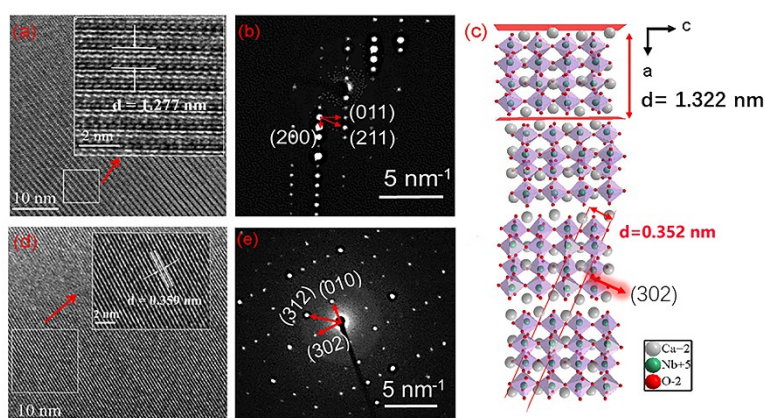


Fig. S2. (a): the HRTEM image and **(b):** the corresponding SAED pattern of the $\text{Ca}_2\text{Nb}_2\text{O}_7$ single crystals of (200) plane. **(d):** the HRTEM image and **(e):** corresponding SAED pattern of $\text{Ca}_2\text{Nb}_2\text{O}_7$ single crystals with (302) crystal plane. **(c):** The crystal structure of $\text{Ca}_2\text{Nb}_2\text{O}_7$ marked with corresponding direction.