

**Achieving high energy efficiency and energy density in PbHfO<sub>3</sub>-based antiferroelectric ceramics**

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Table. S1-1 The permittivity of PLHSx ceramics at room temperature.

| Composition (Hf <sup>4+</sup> ) | 1kHz | 10 kHz | 100 kHz | 1000 kHz |
|---------------------------------|------|--------|---------|----------|
| 0.41                            | 245  | 240    | 237     | 235      |
| 0.43                            | 256  | 253    | 250     | 249      |
| 0.45                            | 280  | 278    | 277     | 277      |
| 0.47                            | 294  | 293    | 292     | 291      |

Table. S1-2 The permittivity of PLHSx ceramics at the temperature of AFE-MCC.

| Composition (Hf <sup>4+</sup> ) | 1kHz | 10 kHz | 100 kHz | 1000 kHz |
|---------------------------------|------|--------|---------|----------|
| 0.41                            | 280  | 278    | 275     | 273      |
| 0.43                            | 299  | 296    | 294     | 293      |
| 0.45                            | 332  | 330    | 329     | 330      |
| 0.47                            | 351  | 350    | 349     | 349      |

Table. S1-3 The permittivity of PLHSx ceramics at the temperature of MCC-PE.

| Composition (Hf <sup>4+</sup> ) | 1kHz | 10 kHz | 100 kHz | 1000 kHz |
|---------------------------------|------|--------|---------|----------|
| 0.41                            | 278  | 257    | 248     | 247      |
| 0.43                            | 290  | 273    | 267     | 266      |
| 0.45                            | 310  | 305    | 302     | 302      |
| 0.47                            | 331  | 325    | 322     | 321      |

Table. S2-1 The dielectric loss of PLHSx ceramics at room temperature.

| Composition (Hf <sup>4+</sup> ) | 1kHz  | 10 kHz | 100 kHz |
|---------------------------------|-------|--------|---------|
| 0.41                            | 0.03  | 0.0129 | 0.0089  |
| 0.43                            | 0.019 | 0.01   | 0.007   |
| 0.45                            | 0.01  | 0.005  | 0.002   |
| 0.47                            | 0.009 | 0.004  | 0.002   |

Table. S2-2 The dielectric loss of PLHSx ceramics at the temperature of AFE-MCC.

| Composition (Hf <sup>4+</sup> ) | 1kHz   | 10 kHz | 100 kHz |
|---------------------------------|--------|--------|---------|
| 0.41                            | 0.0182 | 0.009  | 0.0076  |
| 0.43                            | 0.011  | 0.006  | 0.005   |
| 0.45                            | 0.008  | 0.004  | 0.002   |
| 0.47                            | 0.006  | 0.003  | 0.002   |

Table. S2-3 The dielectric loss of PLHSx ceramics at the temperature of MCC-PE.

| Composition (Hf <sup>4+</sup> ) | 1kHz   | 10 kHz | 100 kHz |
|---------------------------------|--------|--------|---------|
| 0.41                            | 0.086  | 0.042  | 0.013   |
| 0.43                            | 0.0624 | 0.031  | 0.009   |
| 0.45                            | 0.023  | 0.011  | 0.005   |
| 0.47                            | 0.022  | 0.011  | 0.006   |

Table. S3 The average ionic radii of A-site and B-site cations and tolerance factor of PLHSx ceramics.

| Composition (Hf <sup>4+</sup> ) | R <sub>A-site</sub> (Å) | R <sub>B-site</sub> (Å) | Tolerance factor |
|---------------------------------|-------------------------|-------------------------|------------------|
| 0.41                            | 1.4874                  | 0.6982                  | 0.9795           |

|      |        |        |        |
|------|--------|--------|--------|
| 0.43 | 1.4874 | 0.6986 | 0.9794 |
| 0.45 | 1.4874 | 0.6990 | 0.9792 |
| 0.47 | 1.4874 | 0.6994 | 0.9790 |

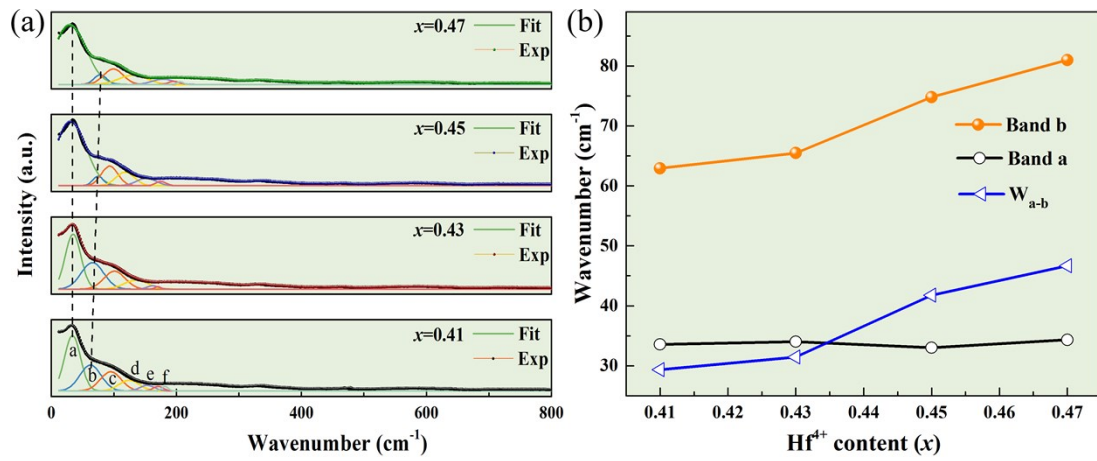


Fig. S1 (a) The Raman spectra and the Lorentzian deconvolutions of PLHS<sub>x</sub> antiferroelectric ceramics; (b) The evolution of position between band a, b and  $W_{a-b}$  ( $W_{a-b}=W_b-W_a$ ) as a function of Hf<sup>4+</sup> content.

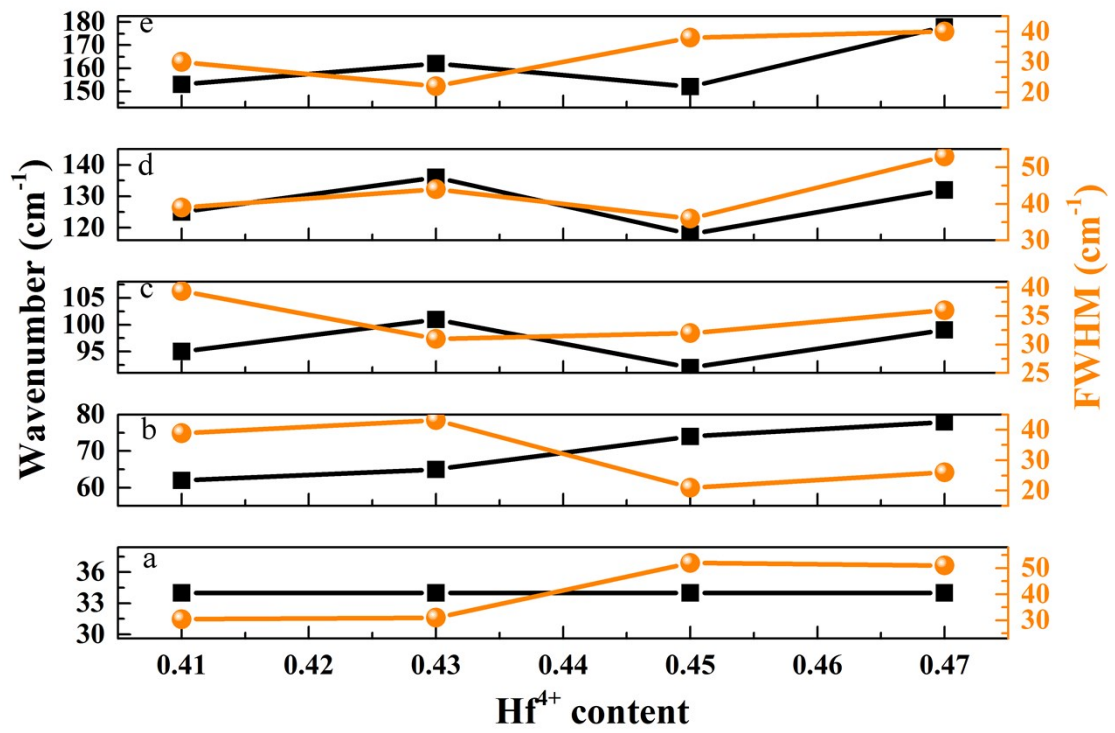


Fig. S2 The temperature dependence of wavenumber and FWHM of modes (a, b, c, d, and e) for PLHSx ceramics.

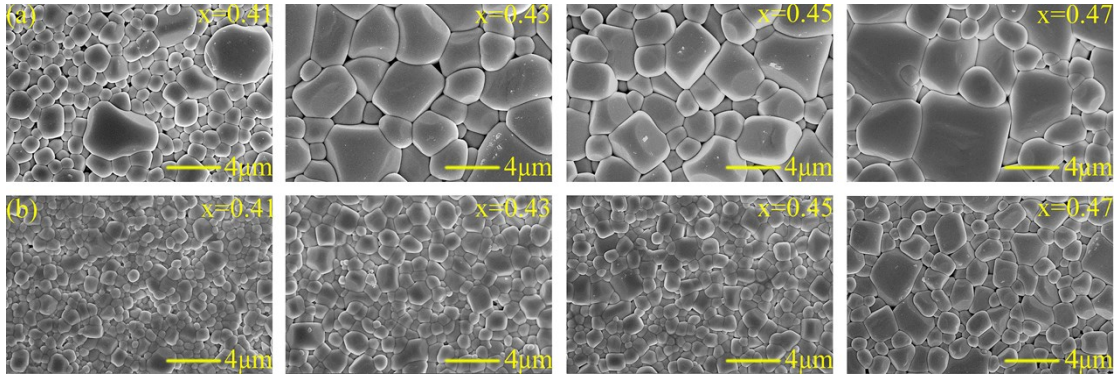


Fig. S3-1 SEM of PLHS<sub>x</sub> ceramics sintered at different temperatures prepared by (a) traditional method (b) rolling process.

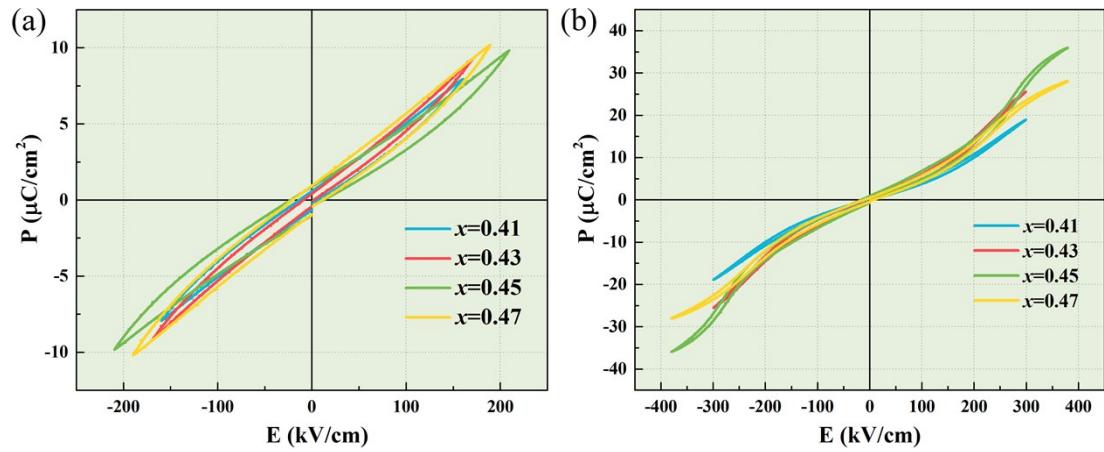


Fig. S3-2 The P-E loops of Pb<sub>0.98</sub>La<sub>0.02</sub>(Hf<sub>x</sub>Sn<sub>1-x</sub>)<sub>0.995</sub>O<sub>3</sub> ceramics made via (a) traditional method and (b) rolling process.

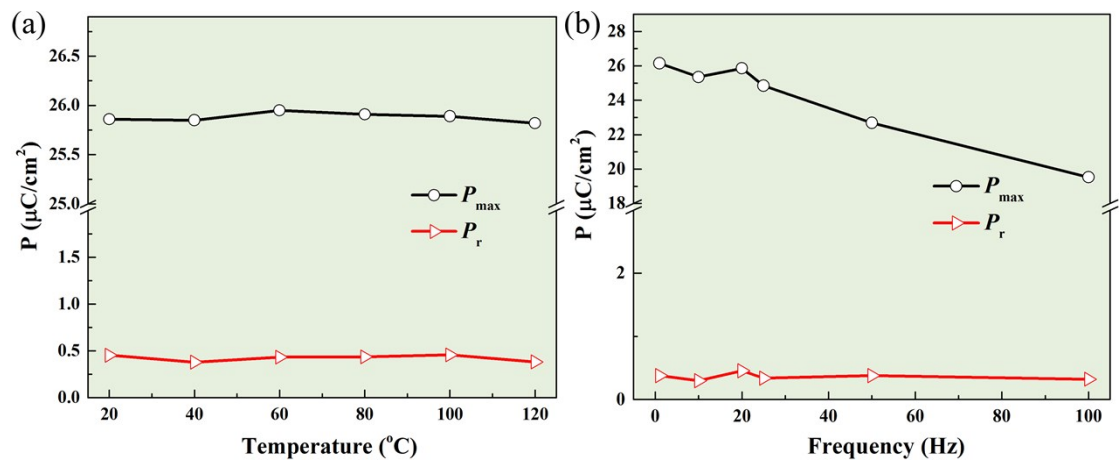


Fig. S4 The  $P_{max}$  and  $P_r$  as a function of (a) temperature and (b) frequency.