Supporting Information

Positive/Negative Electrocaloric Effect Induced by Defect Dipoles in PZT Ferroelectric Bilayer Thin Films

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In this work, the total thickness $l$ of the PZ$_{0.52}$T$_{0.48}$/PZ$_{0.8}$T$_{0.2}$ bilayer thin films is 350 nm, the thickness of the PZ$_{0.52}$T$_{0.48}$ single layer films and the PZ$_{0.8}$T$_{0.2}$ bilayer thin films is about 175 nm. The dielectric constant for the PZ$_{0.8}$T$_{0.2}$ and PZ$_{0.52}$T$_{0.48}$ single layer films is 1068 and 2038, respectively (at 1 kHz). The electric field amplification can be realized in the PZ$_{0.8}$T$_{0.2}$ layer by the uneven dielectric constant of individual layer.1

Fig. S1 The dielectric constant dependence of frequency of the PZ$_{0.8}$T$_{0.2}$ single layer and PZ$_{0.52}$T$_{0.48}$ single layer.

The $P(E)$ loops and $CV$ curves of PZ$_{0.5}$T$_{0.2}$ layer (thickness=175 nm) and PZ$_{0.52}$T$_{0.48}$ layer (thickness=175 nm) are given in Fig. S2. It is consistent well with the previous reported, the double $PE$ loops and the twin-peaks $CV$ curves induced by defect dipoles are observed in in PbZr$_{0.8}$Ti$_{0.2}$O$_3$ single layer films.2
Fig. S2 $P(E)$ loops and CV curves of the PZ$_{0.8}$T$_{0.2}$ single layer and PZ$_{0.85}$T$_{0.15}$ single layer thin film.

(a) $P(E)$ loops of PZ$_{0.85}$T$_{0.15}$ single layer films, (b) $P(E)$ loops of PZ$_{0.8}$T$_{0.2}$ single layer films, (c) CV curves of PZ$_{0.85}$T$_{0.15}$ single layer films, (d) CV curves of PZ$_{0.8}$T$_{0.2}$ single layer films.

In order to further support the existence of the oxygen vacancy, a XPS survey are given in Fig. S3.

Fig. S3 Fig. 1(a) XPS survey spectrum of the PZ$_{0.8}$T$_{0.2}$ individual films and PZ$_{0.85}$T$_{0.15}$/PZ$_{0.8}$T$_{0.2}$ bilayer films, (b) XPS spectrum of O1s for PZ$_{0.8}$T$_{0.2}$ individual films and PZ$_{0.85}$T$_{0.15}$/PZ$_{0.8}$T$_{0.2}$ bilayer films.

REFERENCES
