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]

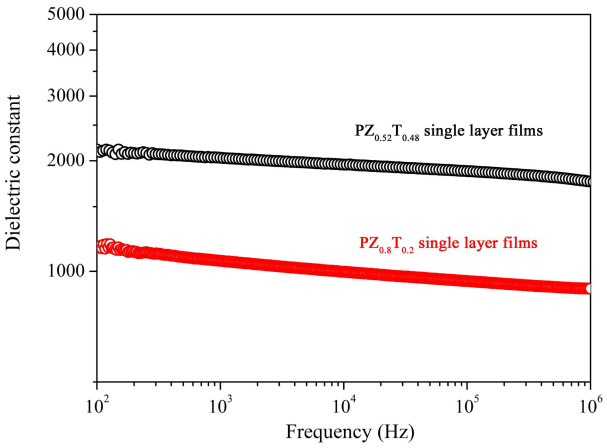

Fig. S1 The dielectric constant dependence of frequency of the $PZ_{0.8}T_{0.2}$ single layer and $PZ_{0.52}T_{0.48}$ single layer.

The *P(E)* loops and *CV* curves of $PZ_{0.8}T_{0.2}$ 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]

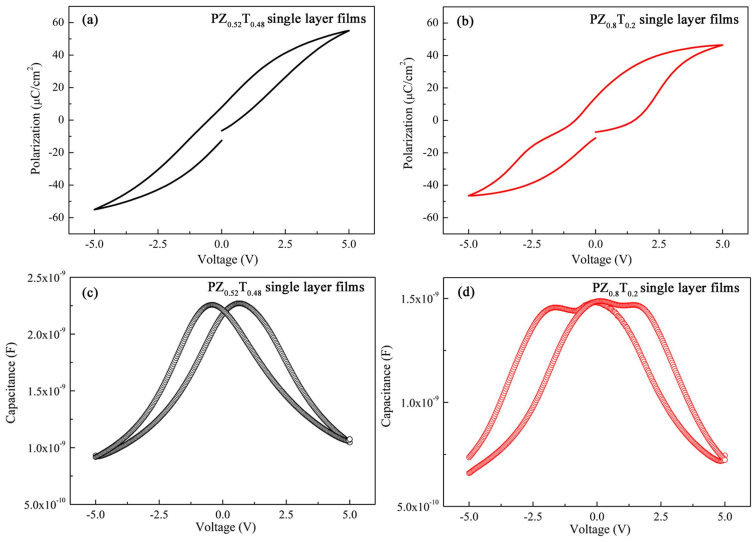

Fig. S2 *P(E)* loops and *CV* curves of the $PZ_{0.8}T_{0.2}$ single layer and $PZ_{0.52}T_{0.48}$ single layer thin film.

(a) *P(E)* loops of $PZ_{0.52}T_{0.48}$ single layer films, (b) *P(E)* loops of $PZ_{0.8}T_{0.2}$ single layer films, (c) *CV* curves of $PZ_{0.52}T_{0.48}$ 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**.

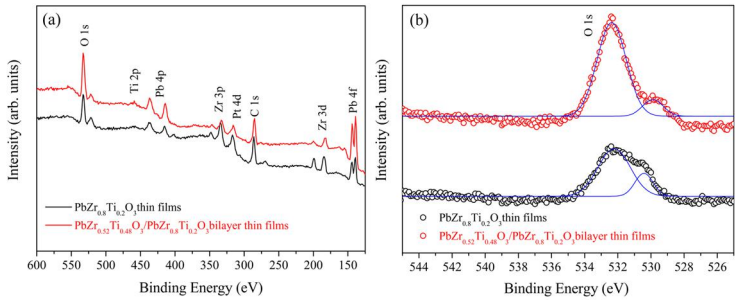

Fig. S3 Fig. 1(a) XPS survey spectrum of the $PZ_{0.8}T_{0.2}$ individual films and $PZ_{0.52}T_{0.48}/PZ_{0.8}T_{0.2}$ bilayer films, (b) XPS spectrum of O1s for $PZ_{0.8}T_{0.2}$ individual films and $PZ_{0.52}T_{0.48}/PZ_{0.8}T_{0.2}$ bilayer films.

REFERENCES

1 T. D. Zhang, W. L. Li, W. P. Cao, Y. F. Hou, Y. Yu, W. D. Fei, *Appl. Phys. Lett.,* 2016, **108**, 162902.

2 Y. T. Pu, J. L. Zhu, X. H. Zhu, Y. S. Luo, M. S. Wang, X. H. Li, J. Liu, J. G. Zhu, D. Q. Xiao, *J. Appl. Phys.,* 2011, **109**, 044102.