## **Electronic Supplementary Information**

## Anisotropic temperature-electric field phase diagrams and domain structure evolutions in rhombohedral Mn-doped PIN-

## **PMN-PT single crystals**

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**Fig. S1** The experimental process of samples preparation for the domain structure observations in Mn:PIN-PMN-PT single crystals. The as-grown crystals were oriented and sliced ((a)-(b)) to the [100]-, [110]- and [111]-oriented samples with three mutually vertical surfaces of (100)/(010)/(001), (001)/(110)/(<sup>110</sup>) and (111)/(<sup>110</sup>)/(<sup>112</sup>) within the pseudocubic coordinates, respectively. And then the crystals with optical surface were polished to a thickness of ~100  $\mu$ m ((c)-(d)). A smooth surface of thin crystals can meet the conditions of domain structure observation under a crossed polarized light microscope.



**Fig. S2** Permissible domain configurations and the 60° domain patterns at 130 °C in [110]-oriented Mn:PIN-PMN-PT single crystals. When electric field is removed, the single-domain state (10) is not stable and partly depolarize into polydomain state. In the [110]-poled crystal analyzed in this work, the four types of 60° domain twins are formed with a dominated [110] domain variant and four adjacent domain variants. The traces of these twin domain walls on (1<sup>1</sup>0) planes form 41° or 139° angles with respect to [001].



**Fig. S3** Permissible domain configurations and the 90° domain patterns at 140 °C in [111]-oriented Mn:PIN-PMN-PT single crystals with 3T engineered domain configuration. The traces of the domain walls on  $(1^{1}0)$  planes form 19.4° or 144.7° angles with respect to  $[11^{2}]$ .



Fig. S4 Unipolar strain-electric field loops for [100]-oriented Mn:PIN-PMN-PT single crystals at various temperatures.



Fig. S5 Unipolar strain-electric field loops for [110]-oriented Mn:PIN-PMN-PT single crystals at various temperatures.



Fig. S6 Unipolar strain-electric field loops for [111]-oriented Mn:PIN-PMN-PT single crystals at various temperatures.