## Supplementary Material

## Giant magnetoelectric effect in perpendicularly magnetized Pt/Co/Ta ultrathin films on

## a ferroelectric substrate

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**Fig. S1** (a) Schematic of the sample structure and the experimental configuration. A Hall bar was patterned to measure AHE. The electric field was applied on the PMN-PT substrate to generate strain via converse piezoelectric effect to modulate the magnetic property of the Pt/Co/Ta ultrathin films; a positive electric field was defined as pointing from PMN-PT to Pt/Co/Ta. (b) AHE curves of Pt/Co/Ta ultrathin films with various thicknesses of Co layer.



**Fig. S2** In situ AHE curves measured under electric fields for perpendicularly magnetized Pt/Co/Ta samples with various Co thicknesses: (a) 1.8 nm, (b) 1.9 nm, (c) 1.95 nm and (d) 2.0 nm. The arrows clearly show that the AHE curves, for all samples, had a remarkable change when applying electric fields. As electric fields increased, the coercive field  $H_C$  in (a) decreased, and the saturated field in (b), (c) and (c) increased, indicating a reduction of PMA by the electric fields.



**Fig. S3** Kerr images taken at zero magnetic field after applying sequential electric fields. The background gradually became darker because of the applied electric field; we subtracted the background to obtain the magnetic domain signal, as shown in Fig. 3.



Fig. S4 Tensile strain along the z axis induced by electric fields, which was estimated based on the shift of the (022) peak in Fig. 4a.



**Fig. S5** Dependence of PMN-PT and bottom Ta interface roughness on electric field, which is deduced from Figure 4b by fitting XRR spectra. In the above figure, we clearly see that the electric field induced a large roughness variation of the PMN-PT substrate and this large substrate roughness variation can be largely suppressed by inserting a 3 nm Ta thin layer.