Electric-field control of non-volatile 180° switching of unidirectional anisotropy field in multiferroic heterostructure

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S1. *M-H* loops were measured at 0° under different electric fields.

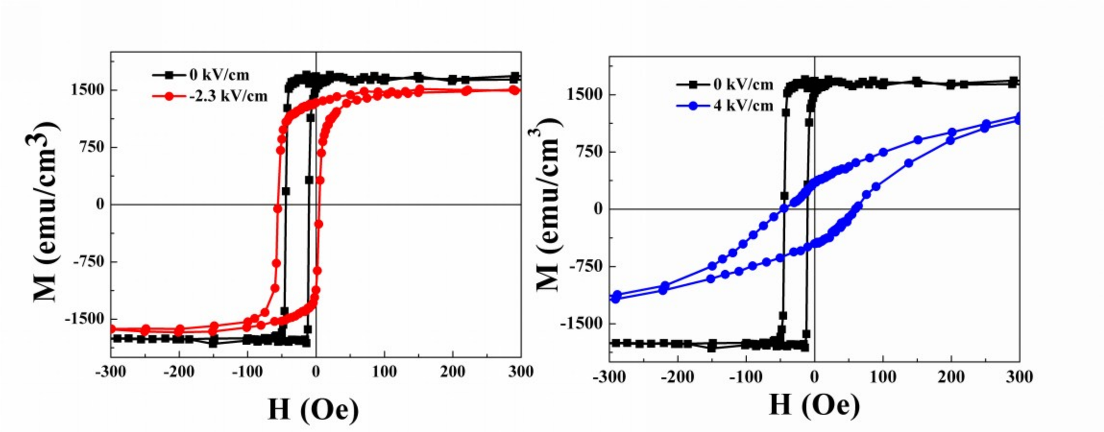

Fig. S1 (a) *M-H* loops measured at 0° under 0 kV/cm and -2.3 kV/cm. (b) *M-H* loops measured at 0° under 0 kV/cm and 4 kV/cm.

The *M-H* loops were measured under -2.3 kV/cm and 4 kV/cm as shown in Fig. S1(a) and (b). The magnetic hysteresis loop show the negative exchange bias field when the electric field at -2.3 kV/cm as shown in Fig. S1(a). However, when the electric field at the 4 kV/cm, the magnetic hysteresis loop show the positive exchange bias field as shown in Fig. S1(b), which show the change of sign of the exchange bias field.

S2. FMR spectra under different electric fields.

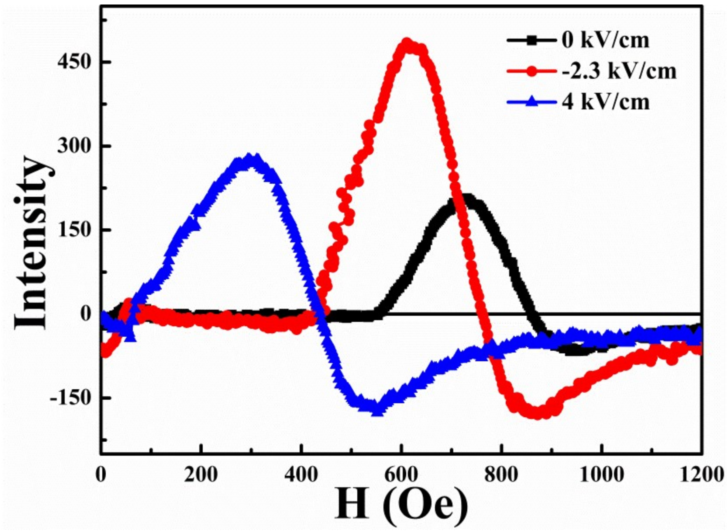

Fig. S2 show the FMR spectra under different electric felds.

As shown in Fig. S2, we use the ferromagnetic resonance (FMR) to measure the magnetization dynamics process, and the change of H_r is relatively large under different electric fields

S3. H_{eb} as function of angle under different electric fields.

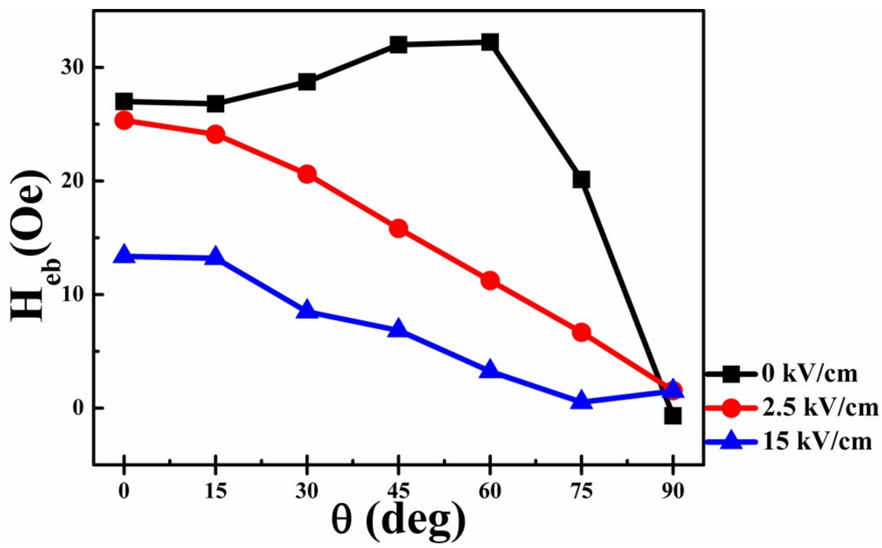

Fig. S3 show the H_{eb} as function of angle under different electric fields

Fig. S3 show the H_{eb} as function of angle under different electric fields, which can better represent the magnitude of the electric field controlling the exchange bias. The control of exchange bias by the electric field is remarkably revealed at θ =60°, which point the exchange bias changes from 32 to 3 Oe under an electric field of +15 kV/cm.

S4. The relative change of angles ϕ and c with electric field with electric field.

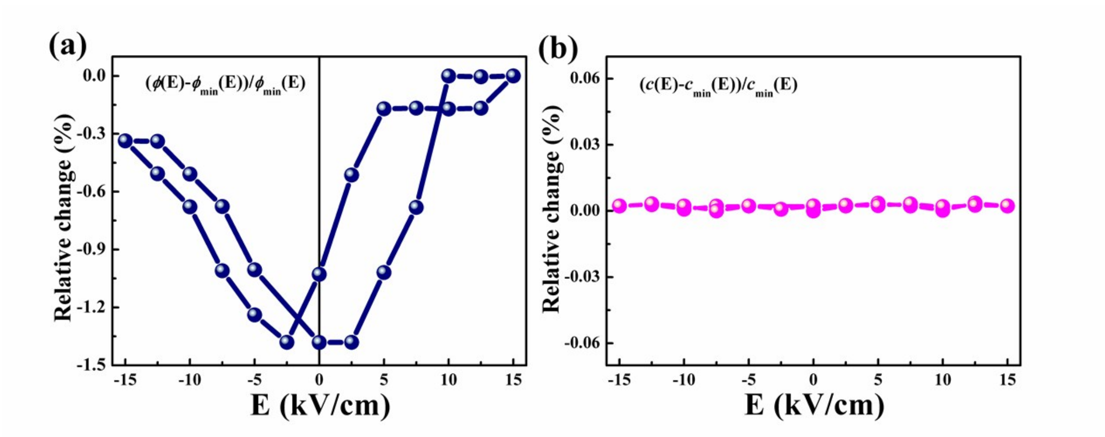

Fig. S4 (a) Shows the relative change (ϕ(E)-ϕ_{min}(E))/ϕ_{min}(E) as a function of applied electric field. (b) the relative change (c(E)-c_{min}(E))/c_{min}(E) as a function of applied electric field.

From the Eq. (4) in the manuscript:

$$H_r = \frac{\left(\frac{2\pi f}{\gamma}\right)^2}{4\pi M_s} + H_{eff}\cos^2(\theta-\phi) + H_{eb}\cos(\theta - c)$$

The phase angle ϕ is the result of the competition between initial uniaxial anisotropies and piezostrain anisotropy. Where the angle c indicates the direction of the unidirectional anisotropy with respect to the initiating point of the measurement. As shown in Fig S4(a), the relative change (ϕ(E)-ϕ_{min}(E))/ϕ_{min}(E) with the electric fields and show exhibits butterfly-like behaviour, the result is consistent with the *S*-*E* curve and can be attributed to the piezo-strain effect. As shown in Fig S4(b), the relative change (c(E)-c_{min}(E))/c_{min}(E) with the electric fields remain almost unchanged with electric fields.