Supplementary Information

Electric-Field-Mediated Magnetic Properties of All-Oxide CoFe₂O₄/

La_{0.67}Sr_{0.33}MnO₃/Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O₃ Heterostructures

Ping Wang,^a Chao Jin,^{*a} Dong Li,^a Yuchen Wang,^a Shasha Liu,^a Xinyue Wang,^a Xin

Pang,^a Dongxing Zheng,^a Wanchao Zheng,^a Rongkun Zheng,^b and Haili Bai^{*a}

^aTianjin Key Laboratory of Low Dimensional Materials Physics and Processing Technology, School of Science, Tianjin University, Tianjin 300350, PR China ^bSchool of Physics, The University of Sydney, NSW 2006, Australia

^{*}Author to whom all correspondence should be addressed.

E-mail: chaojin@tju.edu.cn

E-mail: baihaili@tju.edu.cn



Fig. S1 XRD θ -2 θ pattern of the CFO/LSMO/PMN-PT heterostructures.

The XRD θ -2 θ pattern of the CFO/LSMO/PMN-PT heterostructures is shown in Fig. S1. There are only diffraction peaks of (022), (044) for the CFO layer and (011), (022) for the LSMO layer and PMN-PT substrates.



Fig. S2 (a) AFM and (b) MFM images of the CFO/LSMO/PMN-PT

heterostructures.

The AFM image of the CFO/LSMO/PMN-PT heterostructures is presented in Fig. S2(a). The average roughness 1.09 nm. Figure S2(b) shows the MFM image of the CFO/LSMO/PMN-PT heterostructures. The dark and light contrast areas can be clearly observed, which corresponds to the magnetic domains.

The *M*–*H* curves of the CFO/LSMO/PMN-PT heterostructures along the in-plane [100] direction are given in Fig. S3. The M_R and H_C both increase along the [100] direction under positive and negative electric fields.



Fig. S3 M-H curves of the CFO/LSMO/PMN-PT heterostructures measured under different electric fields along the in-plane [100] direction. The upper-left and bottom-right insets exhibit the enlarged M-H curves.



Fig. S4 *I–V* curves of the LSMO/PMN-PT heterostructures along the in-plane [100] direction measured with electric fields of 0, -10, -0, +10, +0 kV/cm. The upper-left inset shows the enlarged images of the *I–V* curves to clearly distinguish the changes of *I*. The measured voltage from 0 V to 1 V, back to 0 V, then from 0 V to -1 V, back to 0 V in a loop. The upper-right inset shows transport property measurement configuration. The bottom-right inset shows the in-plane *I–V* curve of the CFO layer on PMN-PT substrate measured by Keithley 6487.

The I-V curves of the LSMO/PMN-PT heterostructures along the in-plane [100] direction also present linear characteristic, as shown in Fig. S4. The bottom-right inset gives the linear in-plane I-V curve of single CFO layer on PMN-PT substrate.



Fig. S5 *E*-dependent $\Delta R/R(0)$ curves of the LSMO/PMN-PT heterostructures along the (a) [01-1] and (b) [100] directions.

Figure S5 shows the *E*-dependent $\Delta R/R(0)$ curves of the LSMO/PMN-PT heterostructures. Along both the [01-1] and [100] directions, *E*-dependent $\Delta R/R(0)$ curves exhibit asymmetric butterfly curves, in agreement with the *E*-dependent $\Delta M/M(0)$ curves.



Fig. S6 *E*-dependent $\Delta R/R(0)$ curves of the CFO/LSMO/PMN-PT heterostructures along the out-of-plane [011] direction at (a) the first time, (b) the second time, (c) the third time and (d) the fourth time.

The *E*-dependent $\Delta R/R(0)$ curves of the CFO/LSMO/PMN-PT heterostructures seem like anomalous curves, measured multiple times in Fig. S6. The $\Delta R/R(0)-E$ curves measured at first, second, third and fourth time are not completely same, which properly indicate the complex properties of the CFO/LSMO interfaces.



Fig. S7 *E*-dependent R(E)/R(0) curves of the LSMO/PMN-PT heterostructures with the electric fields of (a), (b) 10 kV/cm and (c), (d) 7.5 kV/cm along the in-plane [01-1] and [100] directions at 300 K, respectively.

The *E*-dependent R(E)/R(0) curves of the LSMO/PMN-PT heterostructures with the electric fields of 10 and 7.5 kV/cm are presented in Fig. S7. The resistances of the LSMO/PMN-PT heterostructures decrease under both the positive and negative electric fields but the reduction is not the same, consistent with *E*-dependent M(E)/M(0) curves.