Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Information for:

Piezoelectrically-Enhanced Exchange Bias in Multiferroic Heterostructures

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Minor hysteresis loops with different film thicknesses of the NFO layers

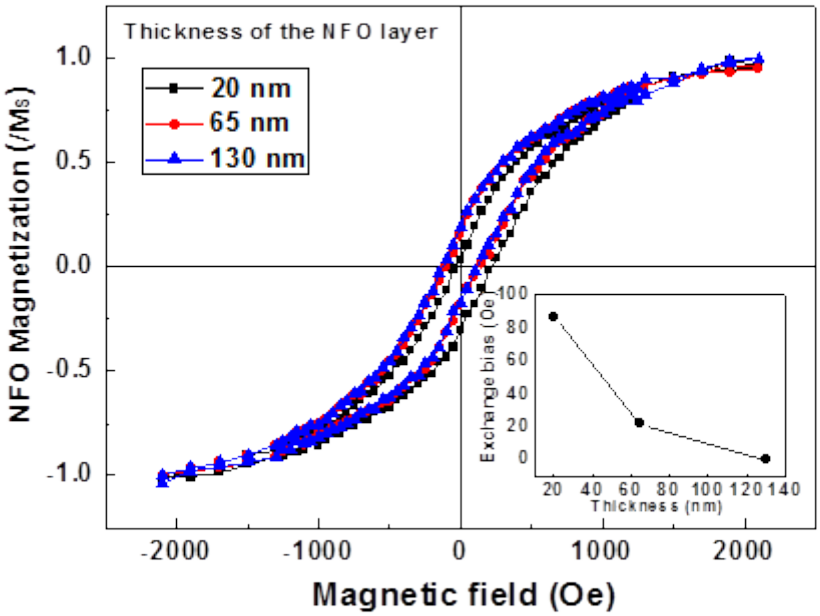

Figure S1. Exchange-biased loops as a function of film thickness of the NFO layer. The samples were initially poled with a magnetic field of -1.2 T to fix the magnetic spins in the CFO layer. The minor hysteresis loop moves back to the original state as the thickness of the soft magnetic layer (NFO) increases, indicating a weakening of the exchange bias effect. Note that this effect is negligible when the NFO thickness is more than 130 nm, as shown in the inset.

The raw data of the minor hysteresis curves

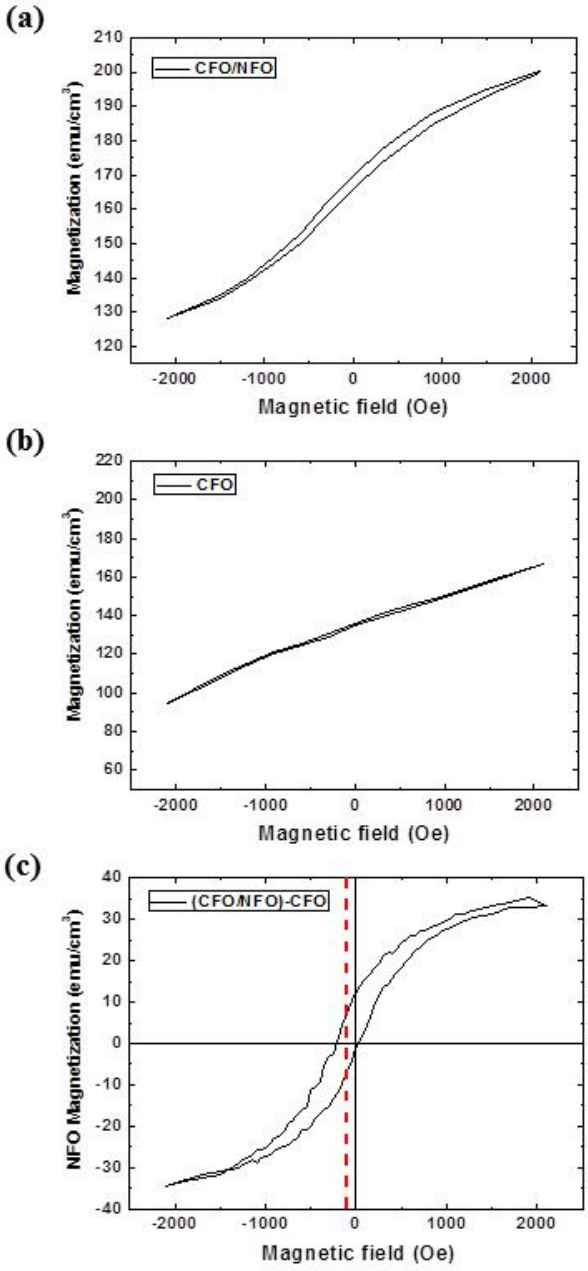

Figure S2. To subtract the CFO contribution from the major hysteresis loop, another CFO reference film on the PMN-PT substrate was grown with the same thickness of 20 nm. Firstly, the CFO reference sample was magnetically poled along the in-plane direction up to +12.5 kOe (in this case). Then, the magnetic hysteresis loop was measured between -2 kOe and +2 kOe in the CFO reference sample. Finally, the minor hysteresis curve was obtained by subtracting the CFO moments from the raw data. (a) raw data before the correction, (b) magnetic moments measured in the CFO/PMN-PT

reference sample between -2 kOe and +2 kOe, (c) the data after the subtraction of (b).

Thickness dependent exchange bias

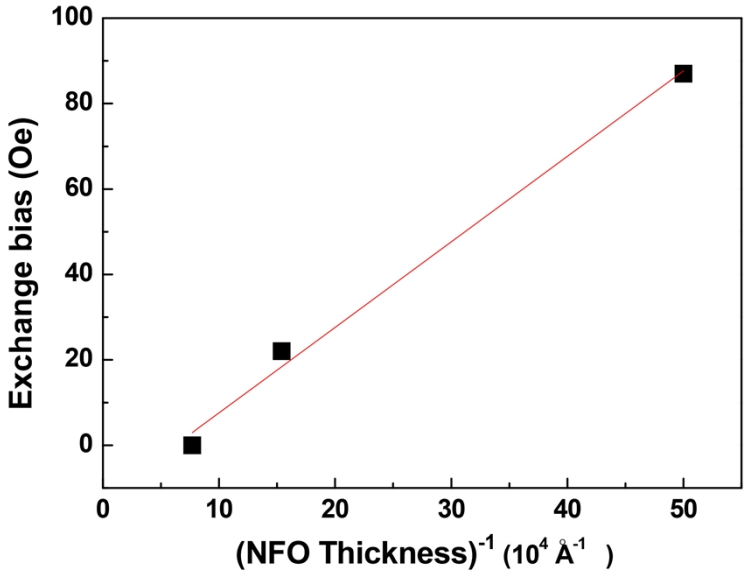

Figure S3. In the NFO/CFO/PMN-PT tri-layer structure, the exchange bias is inversely proportional to the thickness of the soft magnetic layer (NFO), $Hex \propto 1/t_{SM}$. This thickness dependence is clear evidence of the interface spin coupling in the exchange bias effect.[S1,S2]

Polarization-electric field hysteresis loops

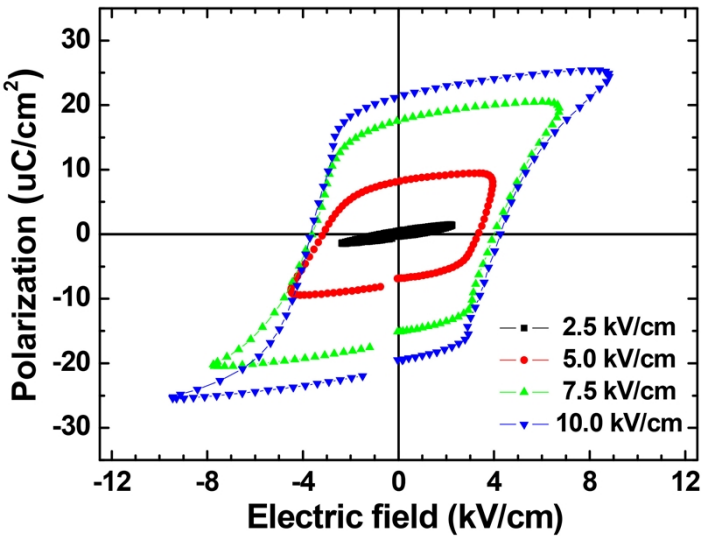

Figure S4. Ferroelectric hysteresis loops measured at 1 kHz in the NFO/CFO/PMN-PT tri-layer structure. Various electric fields up to 10 kV/cm were applied between top and bottom electrodes across the sample. The hysteresis loop shows a well saturated and square-like shape at the electric-field strength of 10 kV/cm (Pr = 21.3 uC/cm^2), suggesting good ferroelectricity, and thus good piezoelectricity.

* References

(S1) Y. Suzuki, R. B. Dover, E. M. Gyorgy, J. M. Phillips, R. J. Felder, *Phys. Rev. B,* 1996, **53**, 14016.

(S2) J. Nogués, J. Sort, V. Langlais, V. Skumryev, S. Suriñach, J. S. Muñoz, M. D. Baró, *Phys. Rep.* 2005, **422**, 65.