Supporting Information

Tuning Ferroelectricity and Ferromagnetism in BiFeO₃/BiMnO₃ Superlattices

Cai Jin,^{a,b} Wanrong Geng,^c Linjing Wang,^b Wenqiao Han,^b Dongfeng Zheng,^a Songbai Hu,^b Mao Ye,^b Zedong Xu,^b Yanjiang Ji,^b Jiali Zhao,^e Zuhuang Chen,^f Gan Wang,^b Yunlong Tang,^c Yinlian Zhu,^c Xiuliang Ma^c and Lang Chen^{*,b}

^aSchool of Physics, Harbin Institute of Technology, Harbin 150081, China.

- ^bDepartment of Physics, Southern University of Science and Technology, Shenzhen 518055, China.
- ^cShenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Wenhua Road 72, 110016 Shenyang, China.
- ^dGuangdong Provincial Key Laboratory of Quantum Engineering and Quantum Materials and Institute for Advanced Materials, South China Academy of Advanced Optoelectronics, South China Normal University, Guangzhou 510006, China.
- ^eLaboratory of Synchrotron Radiation, Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100039, China
- ^fSchool of Materials Science and Engineering, Harbin Institute of Technology, Shenzhen 518055, China.
- * E-mail: <u>chenlang@sustech.edu.cn</u>



Figure S1. AFM scans of the BFO₁/BMO₁ superlattices grown on STO substrates, $R_{\rm rms} \approx 200$ pm.



Figure S2. The φ scans of the BFO/BMO superlattices (202) and STO (202) planes.



Figure S3. Out-of-plane lattice constant c_{pc} of SL-0 as a function of unit cell per period (*m*).



Figure S4. Atomic resolution STEM image in HAADF mode. The uniform contrast

indicates a homogeneous film composition.



Figure S5. Schematic diagram of samples used to measure *P-E* loops. Metallic SRO approximately 20 nm thick serves as the bottom electrode and insulating STO about 5 nm thick deposited on the top and bottom of the superlattices forms a sandwich structure. The Pt top electrodes with the size of 20 β 20 μ m² were fabricated on the surface of the superlattices by photolithography and magnetron sputtering.



Figure S6. Phase of PFM poling map of BFO_{10}/BMO_{10} superlattices at RT. (a) PFM poling map written at +/-8 V. (b) PFM poling map written at -/+8 V.



Figure S7. In-plane field dependent magnetization *M*-*H* hysteresis loops at 10 K for single-layer BFO thin films on (001) STO substrates.



Figure S8. Magnetic properties of single-layer BMO thin films on (001) STO substrates. (a) In-plane field dependent magnetization M-H hysteresis loops at 10 K. (b) In-plane temperature dependent magnetization M-T curves at 1000 Oe.



Figure S9. Magnetization M_S as a function of unit cell per period (*m*).