Supplementary Information

Novel multiferroicity in $GdMnO_3$ thin films with self-assembled nano-twinned domains

Xiang Li[1], Chengliang Lu[2], Jiyan Dai[3], Shuai Dong[4], Yan Chen[3], Ni Hu[5], Guangheng Wu[1], Meifeng Liu[1], Zhibo Yan[1], and Jun-Ming Liu[1,6]

[1]*Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, China,*

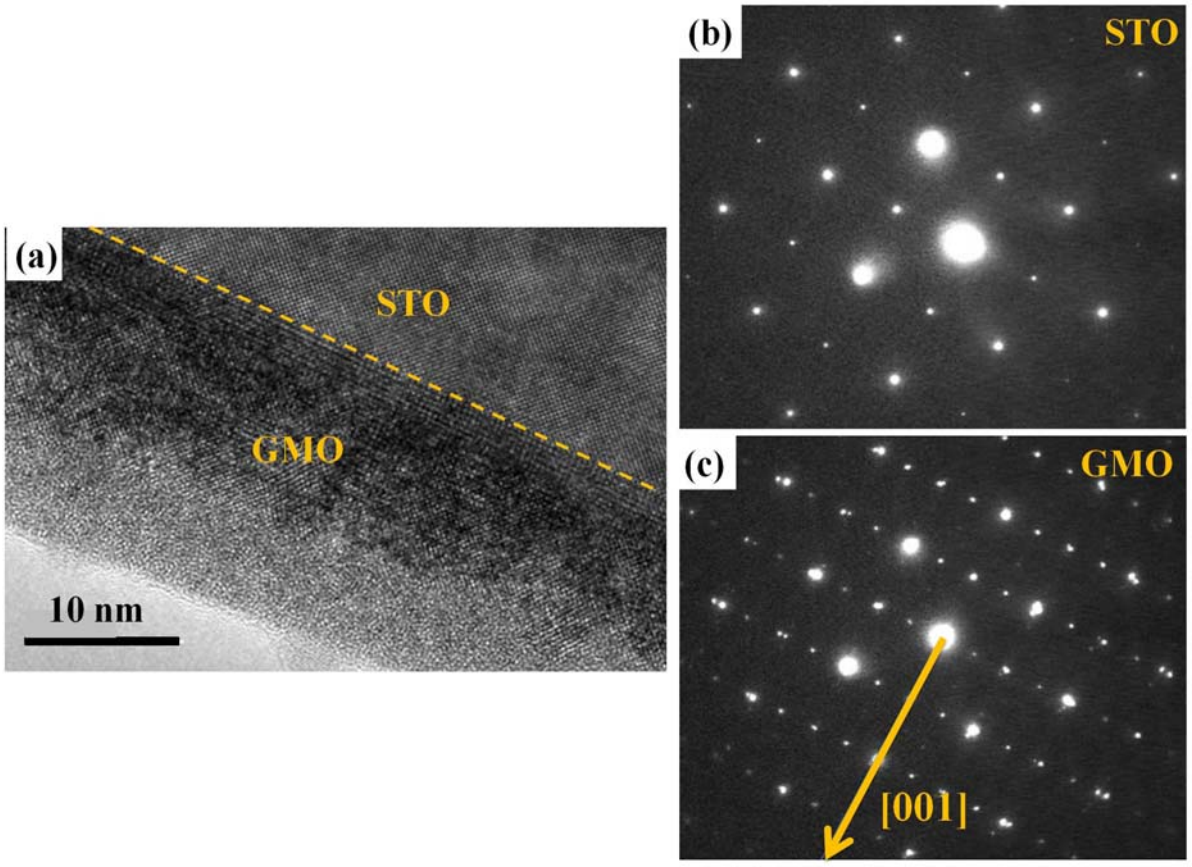
[2]*School of Physics, Huazhong University of Science and Technology, Wuhan 430074, China*

[3]*Department of Applied Physics, Hongkong Polytechnic University, Hongkong, China*

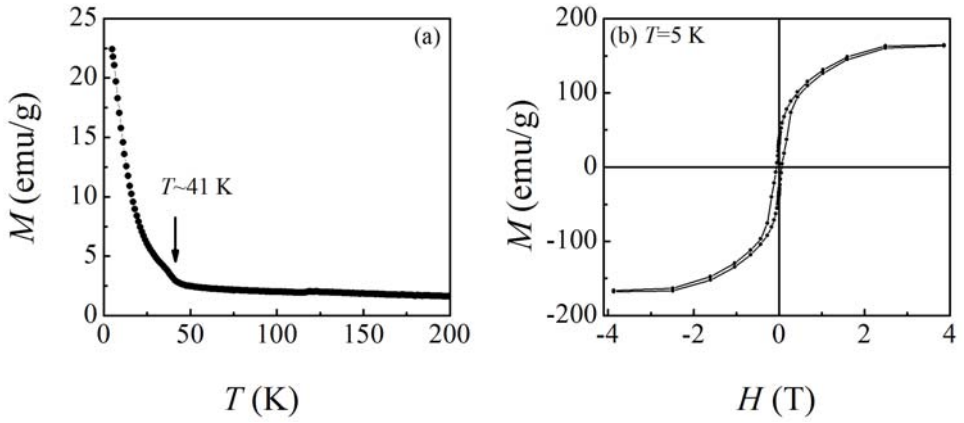
[4]*Department of Physics, Southeast University, Nanjing 211189, China*

[5]*Department of Physics, Hubei University of Technology, Wuhan 430068, China*

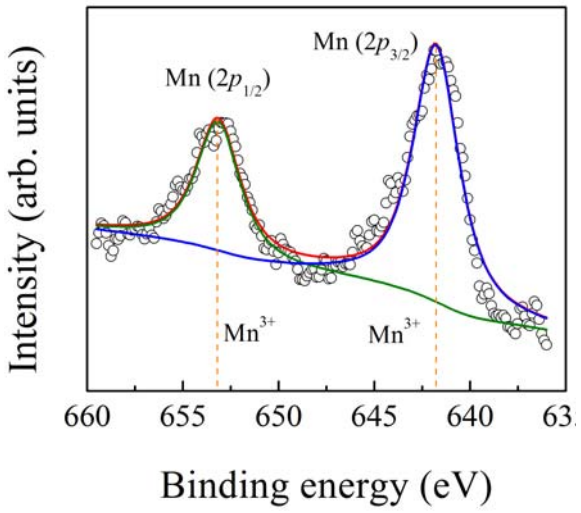
[6]*Institute for Quantum Materials, Hubei Polytechnic University, Huangshi 435000, China*



Supplementary Figure S1: (a) Cross-section TEM of a 10 nm GMO/STO (001) film. Sharp substrate-film interface (indicated by dashed line) can be seen. The corresponding electron diffraction patterns of the film (b) and the substrate (c) show well aligned diffraction spots, confirming the epitaxial growth of the GMO thin film on STO.



Supplementary Figure S2: Magnetization as a function of temperature (a) and magnetic field (b) for the 10 nm thick film. The magnetic properties of the film with t =10 nm are quite similar to that of the film with t =110 nm (shown in Fig. 4 in the main text), including the abrupt increase in *M(T)* curves and the hysteresis in *M(H)* curves. Quantitatively, the film with t =10 nm has a slightly higher onset temperature of the abrupt increase (~41 K), but a smaller coercive field (0.6 kOe) than the film with t =110 nm.



Supplementary Figure S3: XPS spectra for the film with *t*= 110 nm, on which all the measurements were performed. The circles are the experimental data, and the curves are fitting curves. Only Mn^{3+} was detected in the film, which is the same as the case in bulk $GdMnO_3$ crystals.