

Supplementary Information

Stabilization of a honeycomb lattice of IrO_6 octahedra by formation of ilmenite-type superlattices in MnTiO_3

Kei Miura¹, Kohei Fujiwara^{1*}, Kei Nakayama², Ryo Ishikawa^{2,3}, Naoya Shibata² and

Atsushi Tsukazaki^{1,4}

¹*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

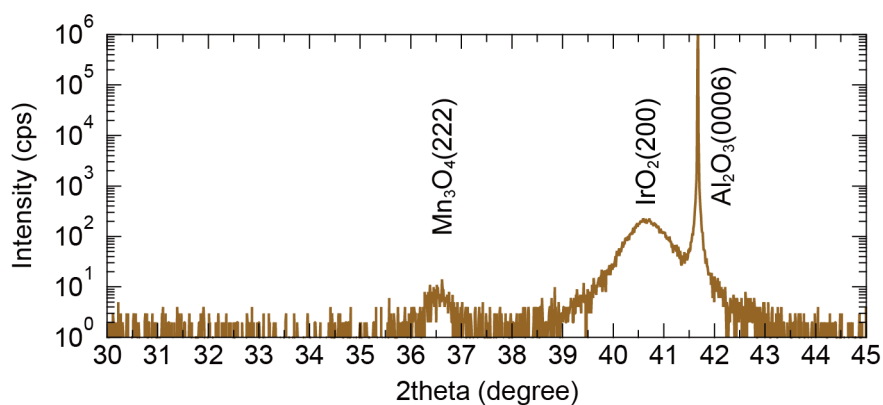
²*Institute of Engineering Innovation, The University of Tokyo, Tokyo 113-8656, Japan*

³*PRESTO, Japan Science and Technology Agency, Kawaguchi 332-0012, Japan*

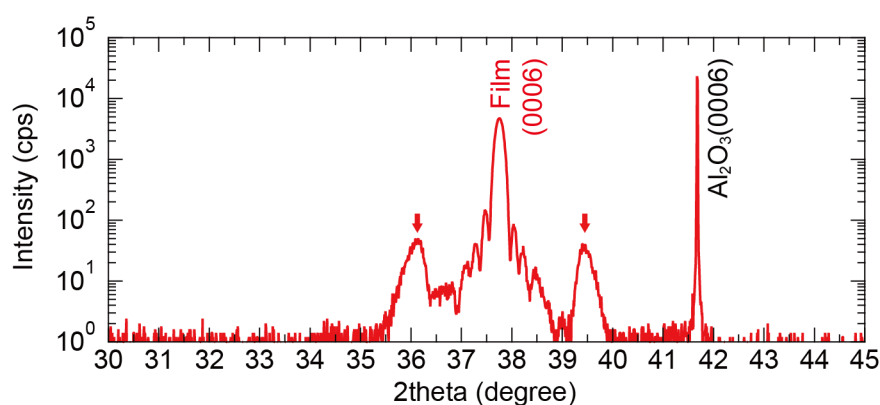
⁴*Center for Spintronics Research Network (CSRN), Tohoku University, Sendai 980-8577, Japan*

* Author to whom correspondence should be addressed: kfujiwara@imr.tohoku.ac.jp

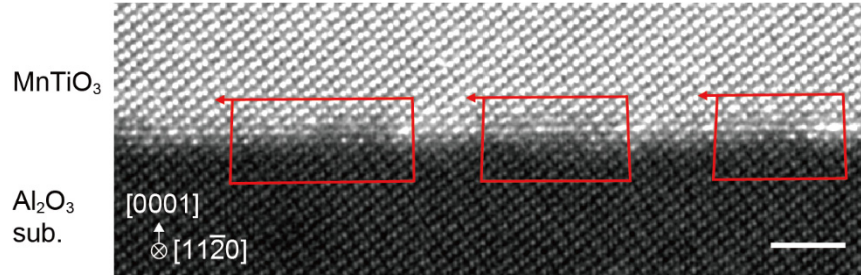
Supplementary Figures



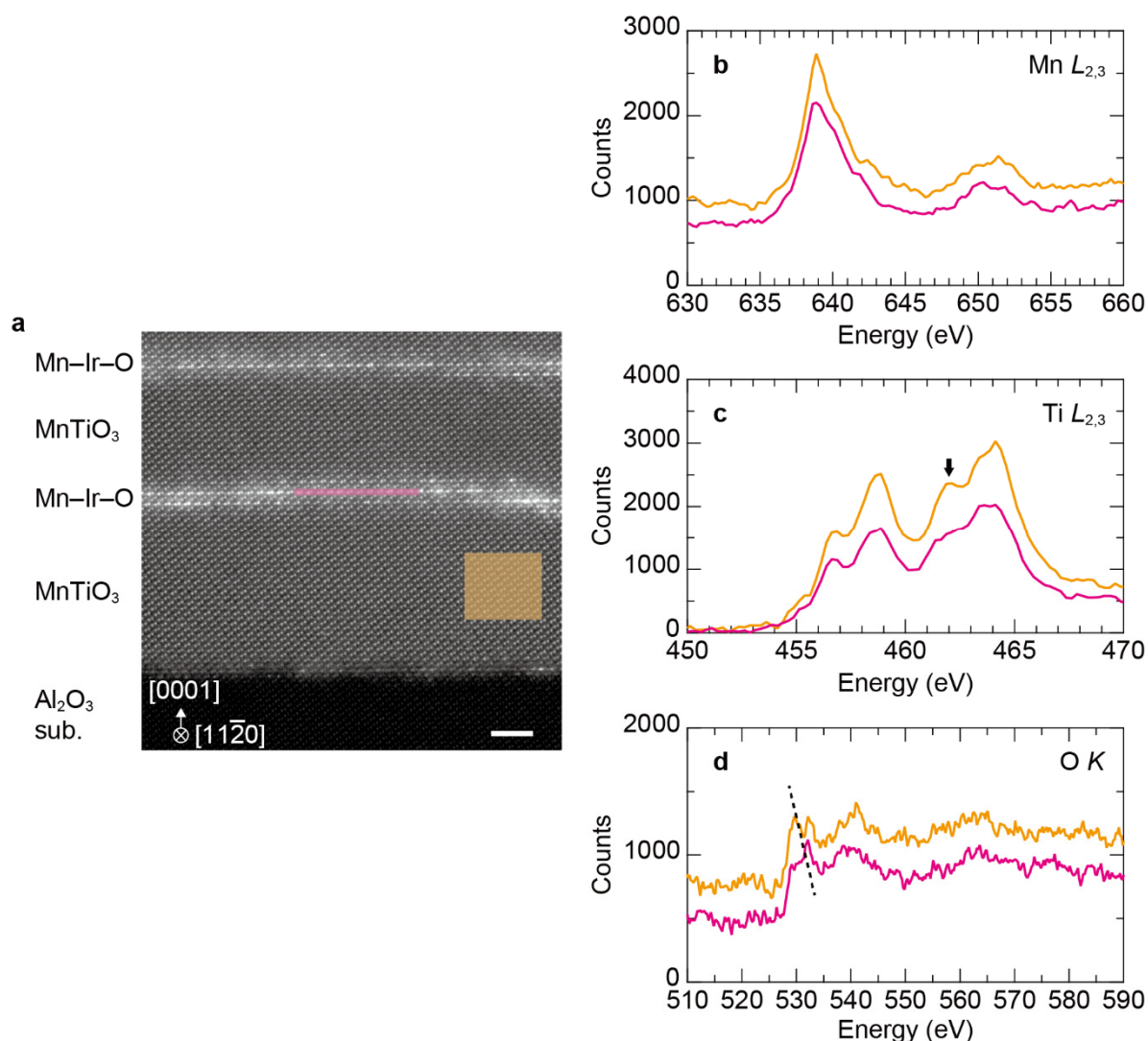
Supplementary Figure 1. Typical out-of-plane XRD pattern of Mn–Ir–O film. The film, with the thickness of approximately 40 nm, was directly deposited on $\text{Al}_2\text{O}_3(0001)$ without a MnTiO_3 buffer layer.



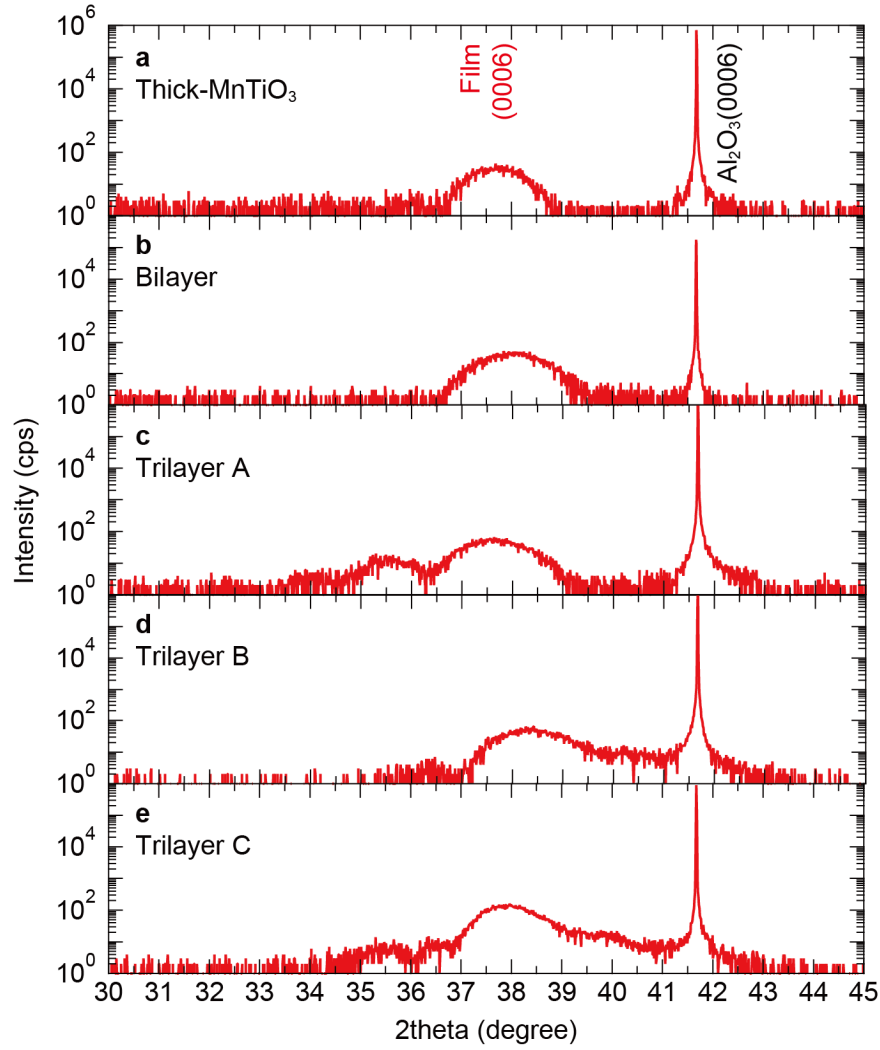
Supplementary Figure 2. Out-of-plane XRD pattern of a $[\text{MnTiO}_3/\text{Mn-Ir-O}]_8/\text{MnTiO}_3(\text{buffer})$ film analysed using STEM. Red arrows indicate superlattice reflections, based on which the single-unit length of the superlattice was calculated to be 5.6 nm. From the thickness fringes around the (0006) peak, the total film thickness was calculated to be 50 nm. These values are in good agreement with those in the HAADF image (Fig. 3a).



Supplementary Figure 3. Atomic-resolution HAADF-STEM image at the heterointerface between MnTiO_3 and the Al_2O_3 substrate. As MnTiO_3 is much heavier than Al_2O_3 , we enhanced the contrast to clearly show geometrical misfit dislocations. The Burgers circuits are represented by red lines. The scale bar was 2 nm. In the sample used for the SMR measurements (Fig. 4a), the overgrown Mn–Ir–O layer is 4.3 nm from the MnTiO_3 buffer/ Al_2O_3 substrate interface. Therefore, it is unlikely that the geometrical misfit dislocations significantly affect the magnetic properties of the Mn–Ir–O₃ layer and the SMR responses, which occur on the top surface of the film.



Supplementary Figure 4. STEM and EELS analysis. **a**, HAADF-STEM image of a $[\text{MnTiO}_3/\text{Mn-Ir-O}]_8/\text{MnTiO}_3$ (buffer) film. The orange and pink areas represent the MnTiO_3 buffer and Mn-Ir-O layer regions analysed by EELS, respectively. The scale bar was 2 nm. EELS data: **b**, $\text{Mn } L_{2,3}$, **c**, $\text{Ti } L_{2,3}$, and **d**, $\text{O } K$. The colour codes are the same as those in **a**. The suppression of the shoulder peak in **c** (indicated by a black arrow) is attributed to the formation of Ti^{3+} . The $\text{O } K$ -edge spectra show a shift to high energy (indicated by a black dashed line), which is indicative of oxygen deficiency near the Mn-Ir-O layer.



Supplementary Figure 5. Out-of-plane XRD patterns of samples used for SMR measurements. **a**, 4.3-nm-thick MnTiO₃ monolayer (thick-MnTiO₃), **b**, 1.0-nm-thick Mn–Ir–O/4.3-nm-thick MnTiO₃ bilayer (bilayer), and **c,d,e**, 1.0-nm-, 1.9-nm-, and 4.3-nm-thick MnTiO₃/1.0-nm-thick Mn–Ir–O/4.3-nm-thick MnTiO₃ trilayer (trilayers A, B, and C, respectively).