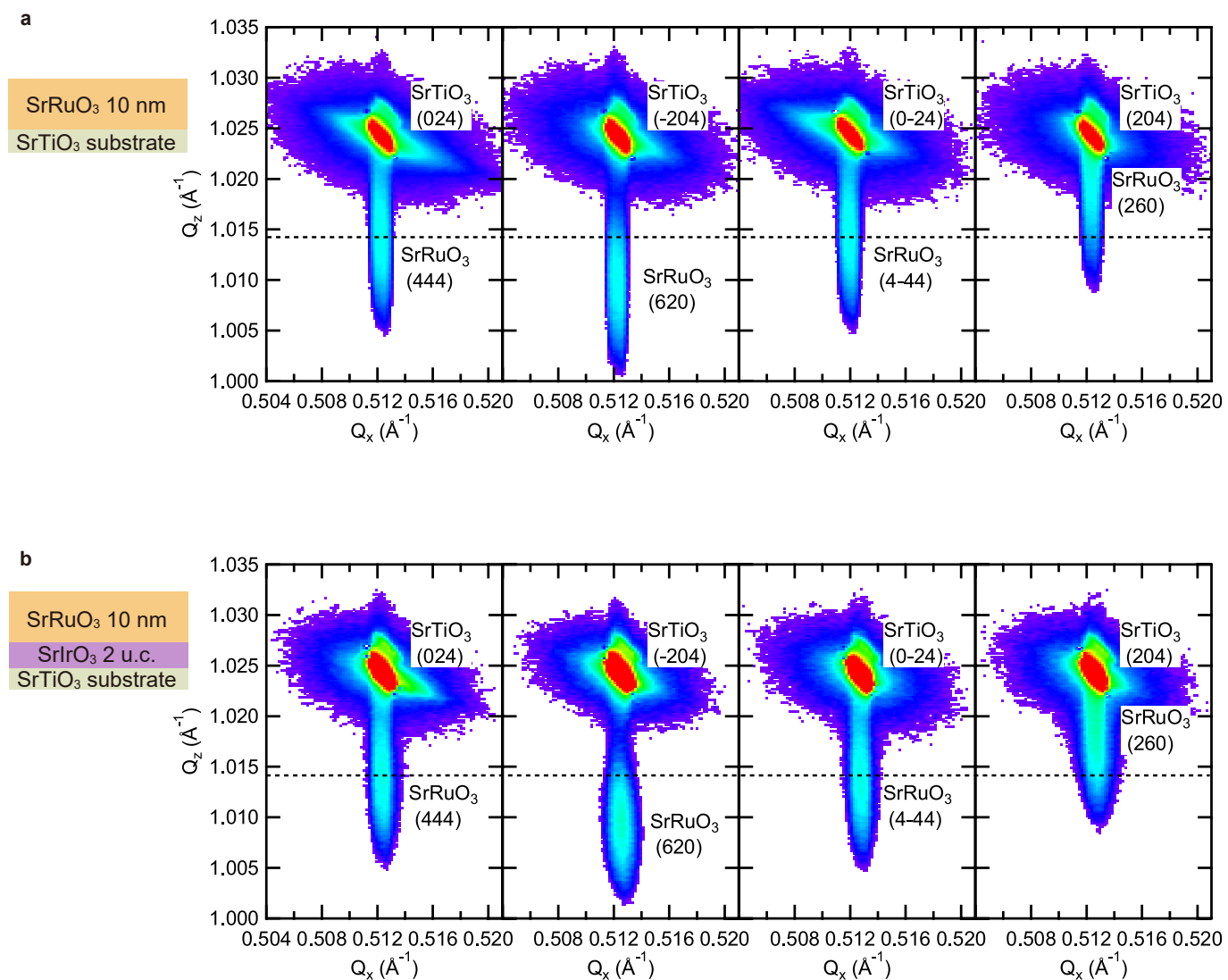
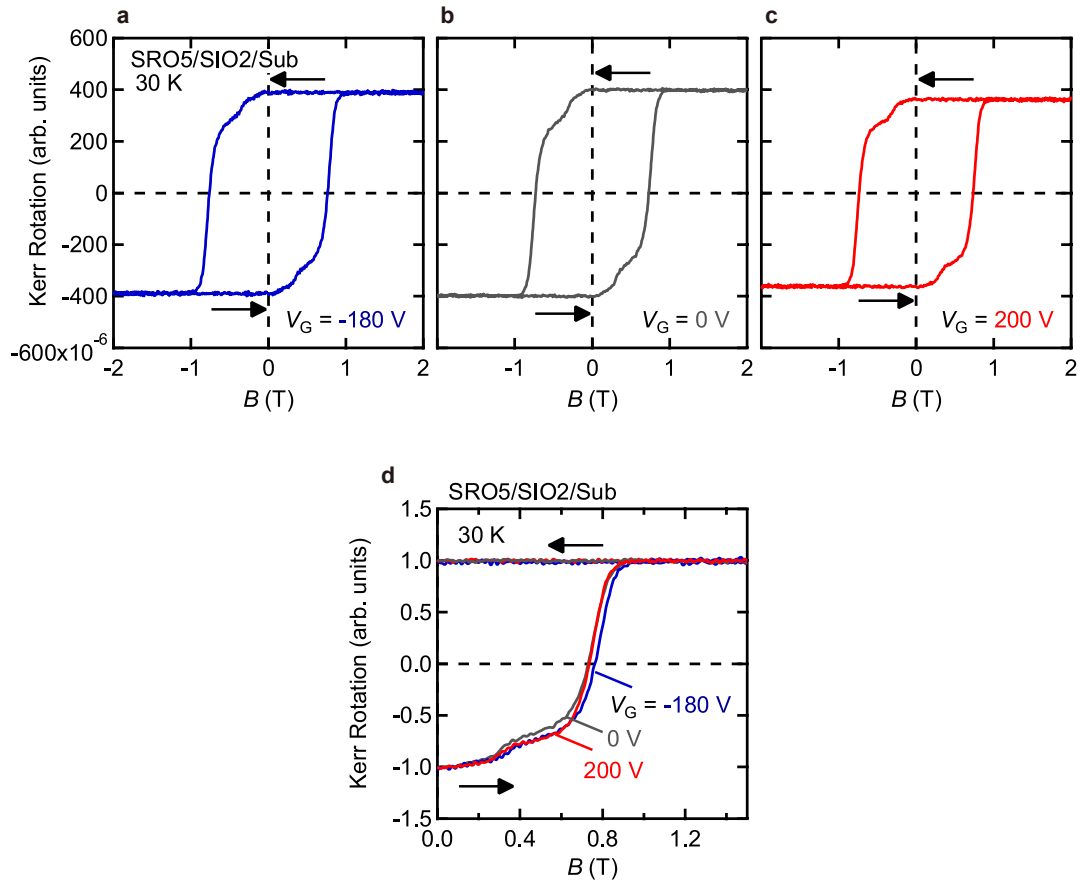




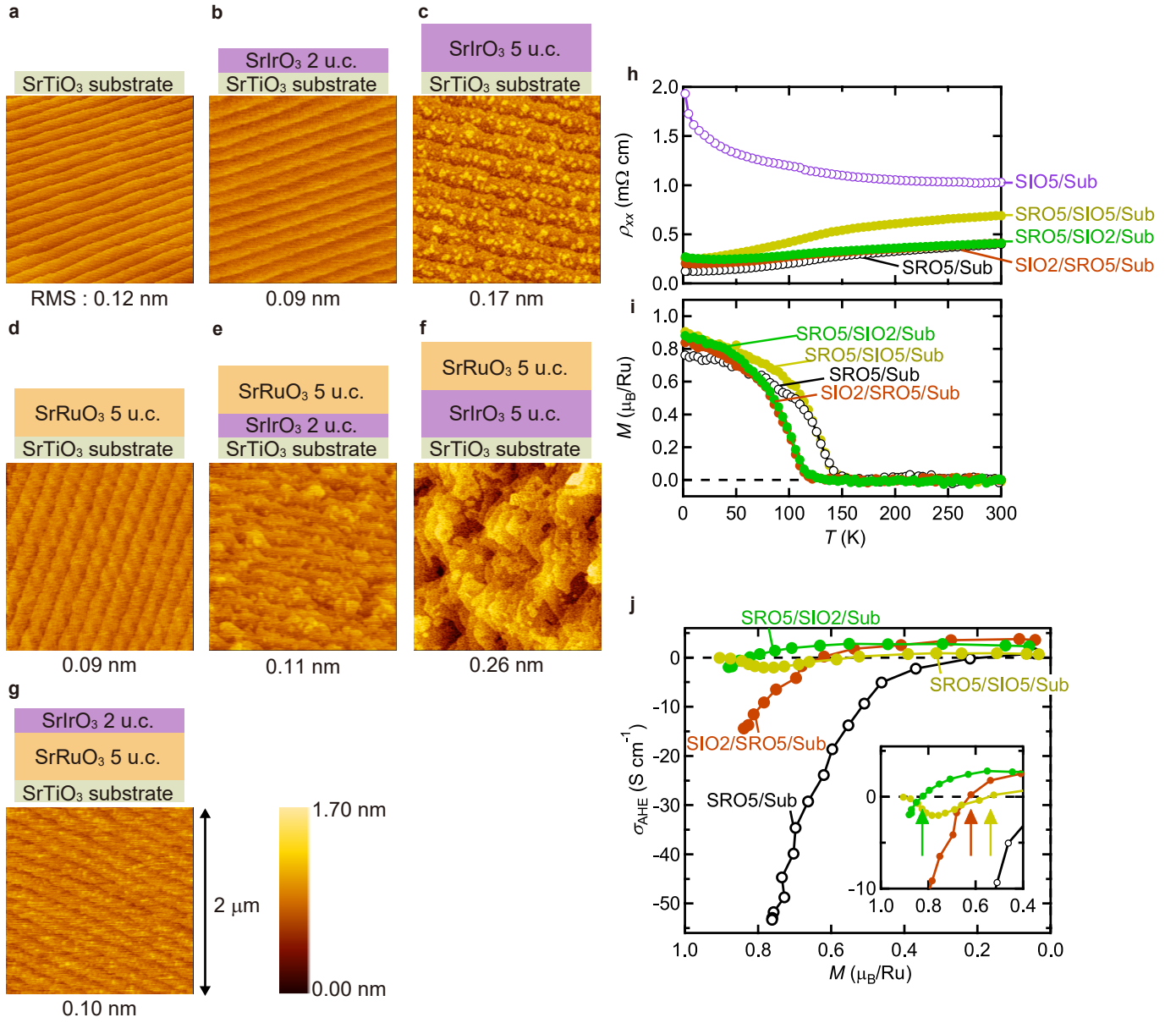
Supplementary Figure 1 | Reciprocal space maps of SrRuO₃ films without/with SrIrO₃ insertion

Reciprocal space maps of 10-nm-thick SrRuO₃ grown on **(a)** a SrTiO₃(001) substrate and **(b)** those with the SrIrO₃ insertion layer. The respective structure is shown in left.



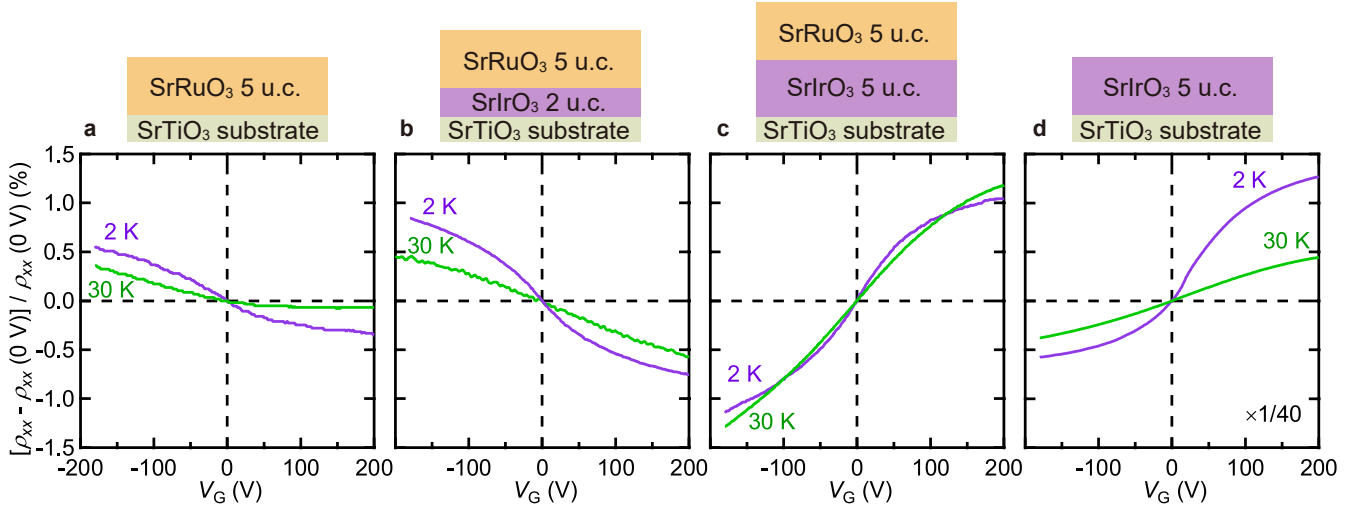
Supplementary Figure 2 | Magneto-optic Kerr rotation as a function of applied electric field

Magnetic-field (B) dependence of Kerr rotation at 30 K under gate voltage $V_G = -180$ V (a), 0 V (b), and 200 V (c) in SRO5/SiO2/Sub. The data are not normalized by ρ_{AHE} in contrast to those shown in Figs. 2a-c in the main text. Black arrows represent field-sweep directions. d, The same Kerr rotation under $V_G = -180$ V (blue), 0 V (grey), and 200 V (red) normalized by the each saturated value at 2 T.



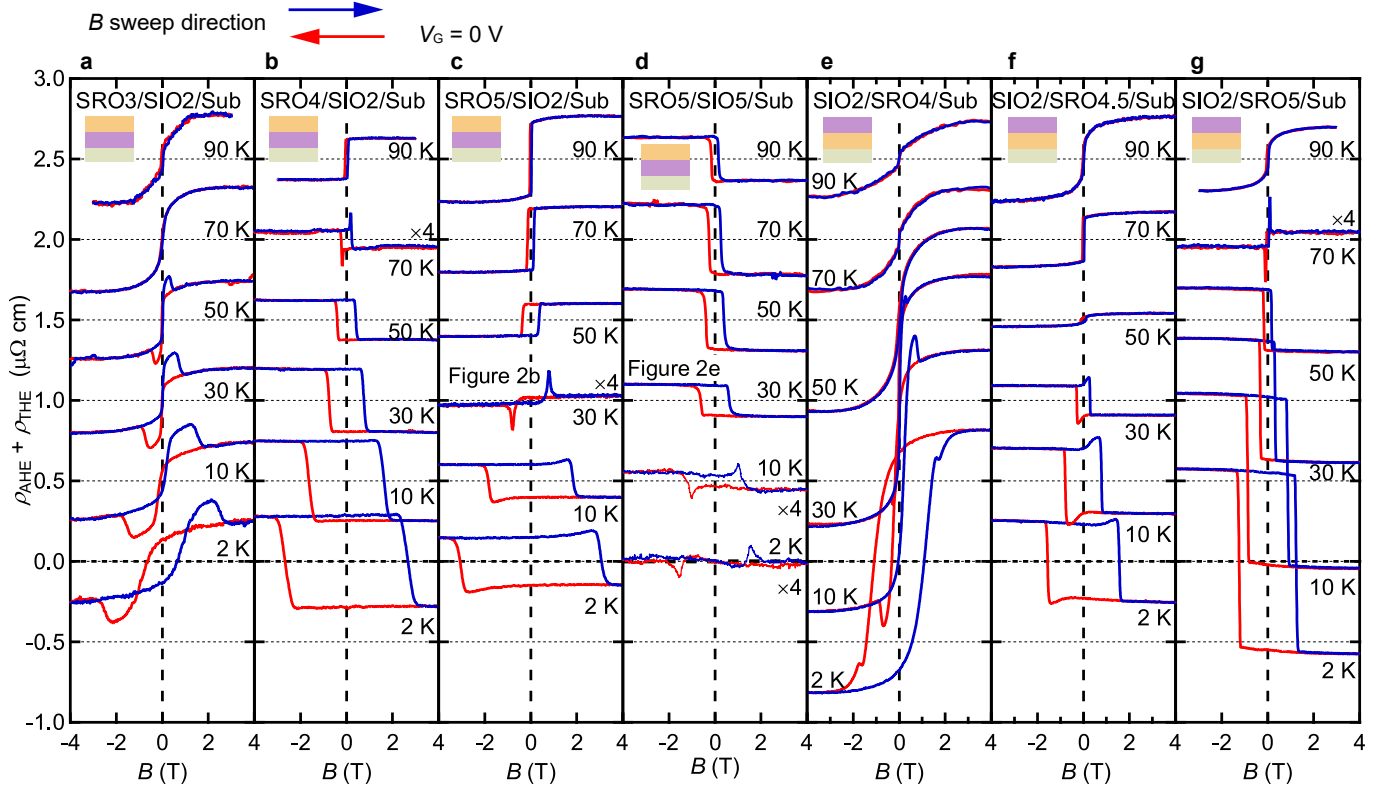
Supplementary Figure 3 | Basic physical properties of SrRuO_3 - SrIrO_3 heterostructures with different SrIrO_3 thickness

AFM images of the surface morphology for films with varied SrIrO_3 thickness (0, 2, 5 unit cells) on SrTiO_3 substrates (**a-c**) and those with a 5-unit-cell SrRuO_3 top layer (**d-f**). The image of 2-unit-cell SrIrO_3 on 5-unit-cell SrRuO_3 is shown for comparison (**g**). Sample structure and Root-Mean-Square (RMS) roughness are shown above and below the images, respectively. Temperature (T) dependence of longitudinal resistivity (ρ_{xx} , **h**) and out-of-plane magnetization measured at 0.1 T (M , **i**). **j**, Anomalous Hall conductivity (σ_{AHE}) as a function of M . Inset shows the magnified view around the sign reversal of σ_{AHE} indicated by arrows.



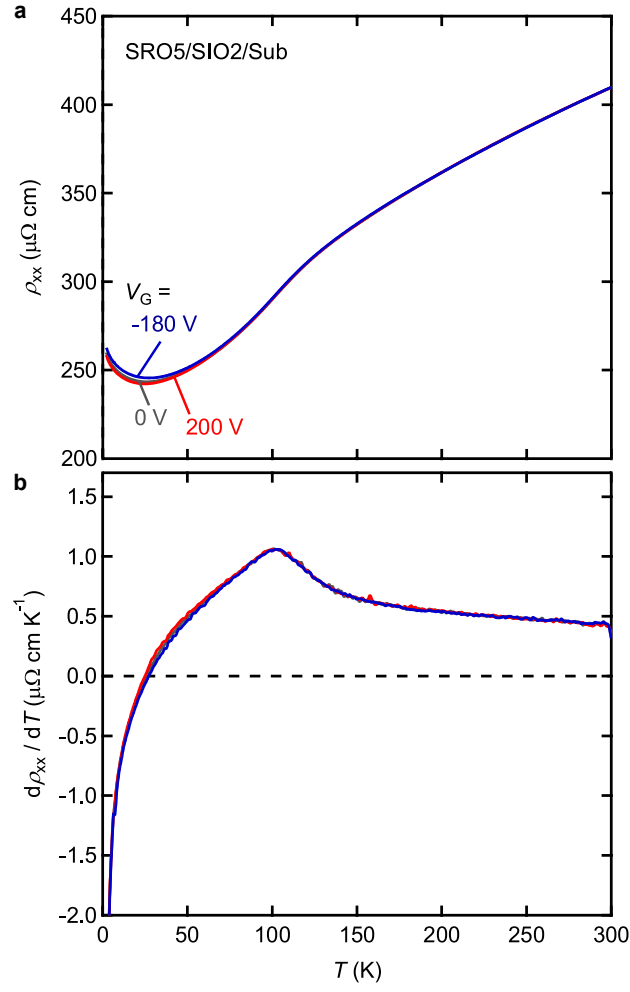
Supplementary Figure 4 | V_G dependence of longitudinal resistivity

a-d, Longitudinal resistivity variation divided by the zero-bias value $[\rho_{xx} - \rho_{xx}(0 \text{ V})]/\rho_{xx}(0 \text{ V})$ as a function of gate voltage (V_G) at 2 K and 30 K. On top, respective sample structure is shown.



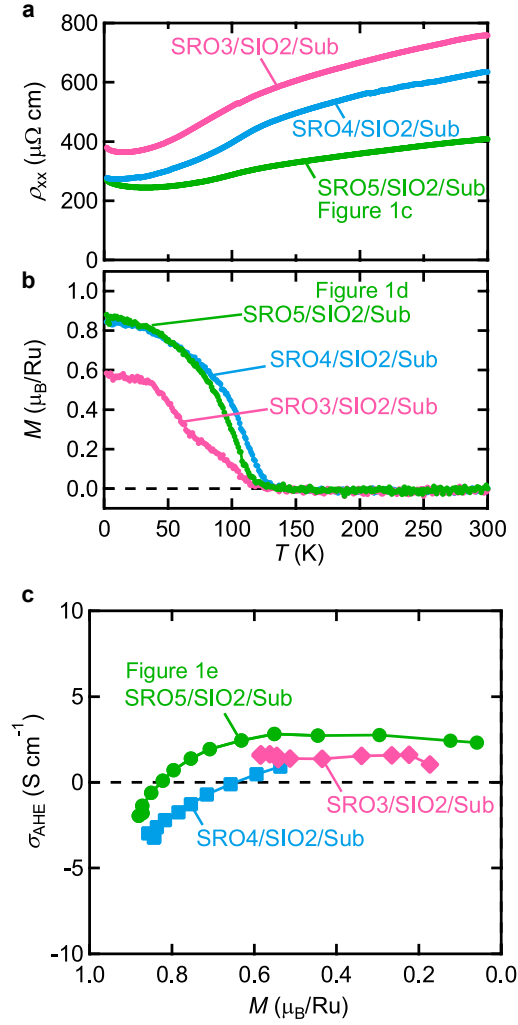
Supplementary Figure 5 | Magneto-transport properties in heterostructures of SrIrO_3 and SrRuO_3

Magnetic-field (B) dependence of anomalous and topological Hall resistivities ($\rho_{\text{AHE}} + \rho_{\text{THE}}$) in $\text{SRO}m/\text{SiO}_2/\text{Sub}$ [$m = 3$ (a), 4 (b), and 5 (c) unit cells], $\text{SRO}5/\text{SiO}_5/\text{Sub}$ (d) and $\text{SiO}_2/\text{SRO}m/\text{Sub}$ [$m = 4$ (e), 4.5 (f), and 5 (g) unit cells] at various temperatures. Stacking sequence is displayed in inset of the each panel. Red (blue) curve corresponds to downward (upward) sweep of B . All measurements were performed under gate voltage $V_G = 0$ V.



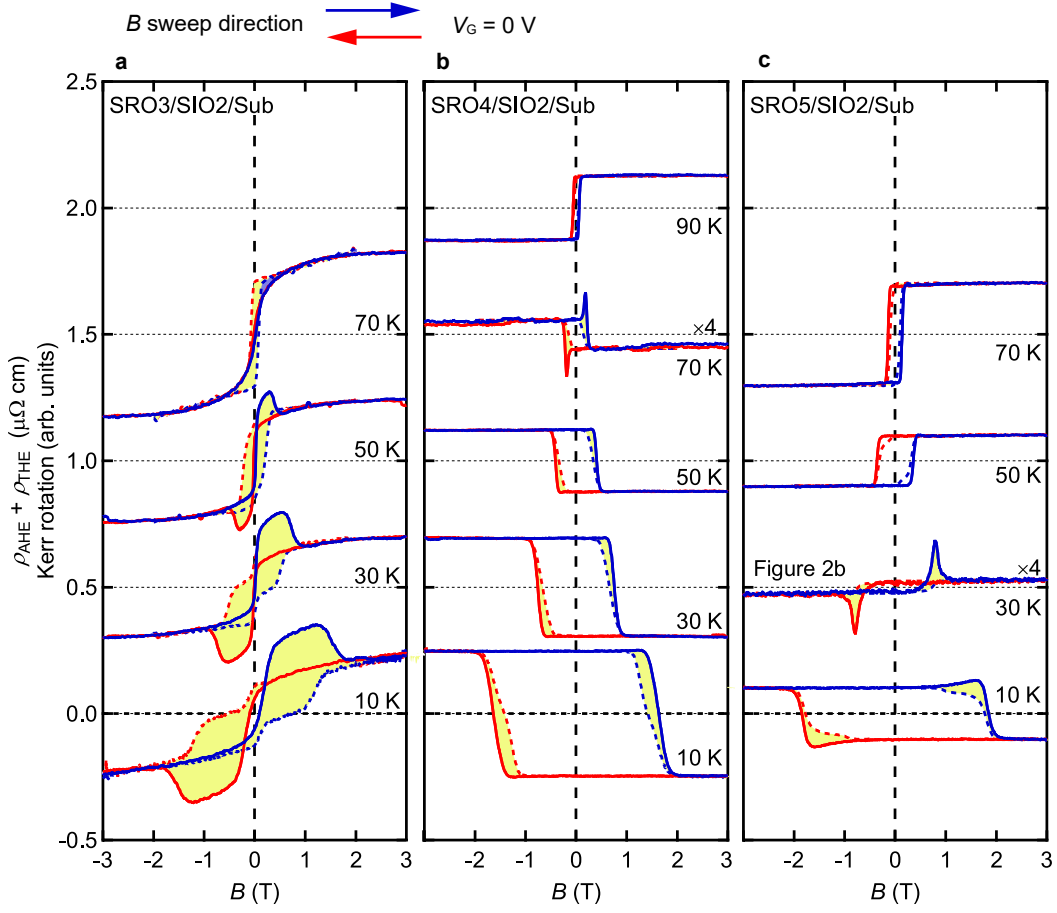
Supplementary Figure 6 | Negligible Curie temperature variation by applied electric field

Temperature (T) dependence of longitudinal resistivity (ρ_{xx} , **a**) and differential resistivity ($d\rho_{xx} / dT$, **b**) under gate voltage $V_G = -180 \text{ V}$ (blue), 0 V (grey), and 200 V (red) in SRO5/SiO2/Sub.



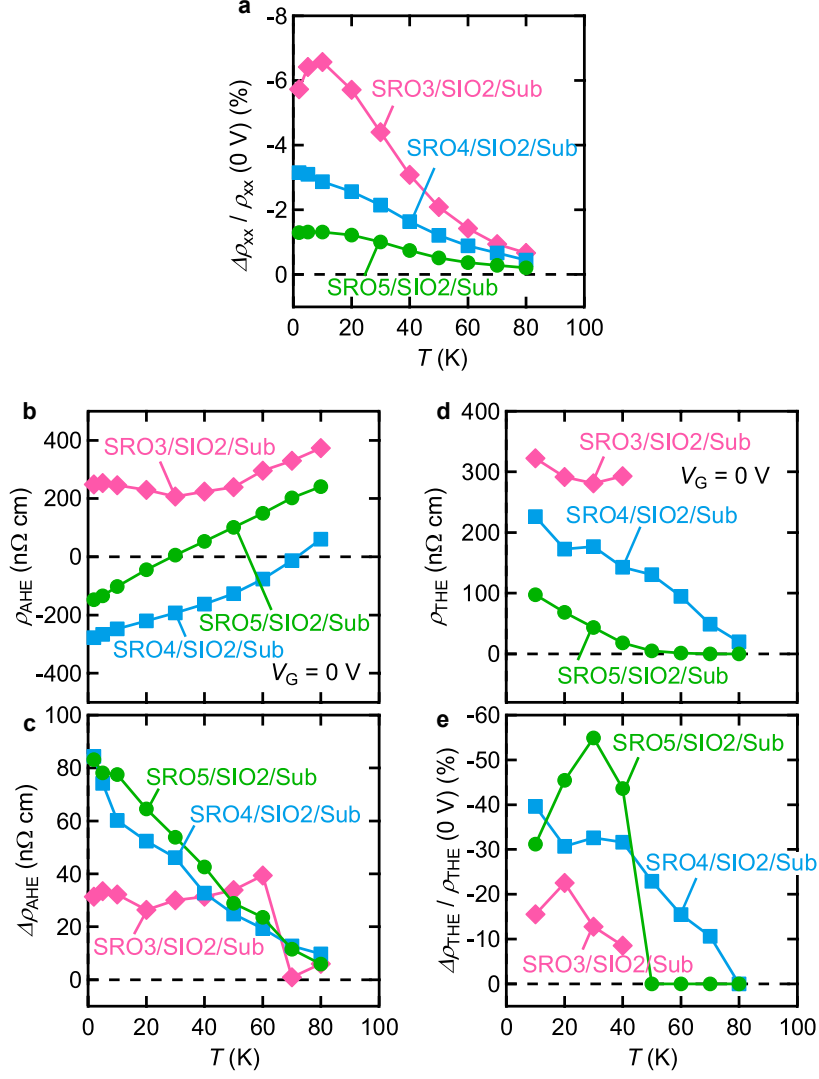
Supplementary Figure 7 | Basic physical properties of SrRuO₃/SrIrO₃/SrTiO₃ with different SrRuO₃ thickness

Temperature (T) dependence of longitudinal resistivity (ρ_{xx} , **a**) and out-of-plane magnetization measured under 0.1 T (M , **b**) in SRO m /SIO₂/Sub ($m = 3, 4$, and 5 unit cells). **c**, Anomalous Hall conductivity (σ_{AHE}) as a function of magnetization (M).



Supplementary Figure 8 | Topological Hall effect deduced from transport and magneto-optic Kerr effect measurements in SRO m /SiO $_2$ /Sub ($m = 3, 4$, and 5)

Magnetic-field (B) dependence of anomalous and topological Hall resistivities ($\rho_{\text{AHE}} + \rho_{\text{THE}}$, solid lines) under gate voltage $V_G = 0$ V for SRO m /SiO $_2$ /Sub [$m = 3$ (a), 4 (b), and 5 (c)]. Kerr rotations are also shown by broken lines. Yellow coloured regions correspond to topological Hall resistivity (ρ_{THE}). Red (blue) curves correspond to downward (upward) sweeps of B .



Supplementary Figure 9 | Temperature dependence of transport properties and their gate modulations in SRO_m/SiO₂/Sub ($m = 3, 4$, and 5)

a, Temperature (T) dependence of variation in longitudinal resistivity normalized by the zero-bias ($V_G = 0$ V) value [$\Delta\rho_{xx}/\rho_{xx}(0\text{ V})$]. T dependence of anomalous Hall resistivity (ρ_{AHE} , **b**), and its modulation amplitude [$\Delta\rho_{\text{AHE}} = \rho_{\text{AHE}}(V_G = 200\text{ V}) - \rho_{\text{AHE}}(V_G = -180\text{ V})$, **c**]. T dependence of topological Hall resistivity (ρ_{THE} , **d**), and its modulation amplitude normalized by zero-bias value [$\Delta\rho_{\text{THE}}/\rho_{\text{THE}}(0\text{ V})$, **e**].

Supplementary Note 1 | Lattice structure of SrIrO₃-inserted SrRuO₃

It has been reported that SrRuO₃ has the structural deformation from its original orthorhombic structure to tetragonal one due to the octahedral rotation mismatch with a buffer layer on a SrTiO₃ substrate¹, accompanying the modification of magnetic anisotropy. In our case of SrRuO₃/SrIrO₃/SrTiO₃, this kind of structural phase change might be triggered since SrIrO₃ can serve as a buffer layer. However, in the structural point of view, the insertion of SrIrO₃ is expected to have negligible effect on the structural properties of the top SrRuO₃ layer. The large structural distortion in the heterointerface of perovskite oxides is known to be induced by the combinations with a large difference in ionic radii, because this leads to a large difference in the distortion of the octahedra formed with *B*-site transition metals and surrounding oxygens. In our case, the ionic radius of Ru⁴⁺ is about 62 pm, which is close to that of Ir⁴⁺ (62.5 pm).

In fact, as shown in the x-ray diffraction data in Supplementary Figs. 1a and 1b, we confirmed that the 10-nm-thick SrRuO₃ films have the same orthorhombic (or lower symmetry) structure at room temperature regardless of the insertion of 2-unit-cell SrIrO₃ on SrTiO₃ substrates, indicating SrIrO₃ has a negligible impact on the lattice structure of SrRuO₃. Furthermore, as shown in Fig. 1d in the main text, the ferromagnetic properties such as Curie temperature (T_C) and the value of magnetization are quite similar between SRO5/SIO2/Sub and SIO2/SRO5/Sub. These similarities are in agreement with the identical lattice structure although we cannot resolve the reciprocal space maps in such thin heterostructures. Therefore we conclude that the stacking order brings a negligible impact on the structural details of SrRuO₃.

Supplementary Note 2 | Magneto-optic Kerr effect (MOKE) under applied electric fields

In Supplementary Figs. 2a-c, we show magnetic-field dependences of Kerr rotation in SRO5/SIO2/Sub at 30 K under different gate biases (V_G). The sign of all measurements at saturation field is the same with each other in spite of the sign inversion of anomalous Hall resistivity (ρ_{AHE}). This fixed sign of Kerr rotation indicates that the magnetization itself is not inverted by the applied electric field, confirming the variation of the proportionality factor R_S in the intrinsic anomalous Hall effect (AHE). We note that the saturated Kerr rotation under each applied electric field has a limited accuracy; this fluctuation is at most 10% of the saturated values at $V_G = 0$ V. Hence, the electric-field-induced variation of magnetization is under this order of magnitude. The same Kerr rotations normalized by each saturated value at 2 T are shown in Supplementary Fig. 2d. The coercive force (H_C) is shifted from 0.76 T ($V_G = 200$ V) to 0.73 T ($V_G = -180$ V) at 30 K; this shift of H_C implies the modulation of spin-orbit coupling (SOC) because the magnetic anisotropy results from SOC.

Supplementary Note 3 | Thickness dependence of SrIrO₃ in SrRuO₃/SrIrO₃/SrTiO₃

In Supplementary Figs. 3a-c, we show atomic force microscopy (AFM) images of the surface morphology for the SrIrO₃ films grown on SrTiO₃(001) substrates (SIO x /Sub, $x = 0, 2, 5$ unit cells, respectively). The clear step-and-terrace structure starts to be degraded in SIO5/Sub, while the structure is kept in SIO2/Sub. This thickness dependence of the surface morphology results from the lack of step-flow growth in SIO x /Sub when x is larger than 2. Supplementary Figures 3d-f show AFM images of SRO5/Sub, SRO5/SIO2/Sub and SRO5/SIO5/Sub respectively. In accord with the trend

of SIOx/Sub, the heterostructure with thicker SrIrO₃ (SRO5/SIO5/Sub) has the worse surface flatness, where the step-and-terrace structure almost disappears. The root-mean-square (RMS) roughness is 0.26 nm, which deviates from the ideal value of a flat step-and-terrace surface of SrRuO₃ (0.11 nm). In Supplementary Fig. 3g, we show the preserved step-and-terrace surface morphology of SIO2/SRO5/Sub for comparison.

Supplementary Fig. 3h shows temperature dependence of longitudinal resistivity (ρ_{xx}) in the samples with 5-unit-cell SrRuO₃. For comparison, we plot ρ_{xx} in SIO5/Sub, indicating the semimetallic temperature dependence as reported in the thicker films^{2,3}. Due to the finite contribution of the SrIrO₃ layer, ρ_{xx} in SRO5/SIO5/Sub is higher than those in other samples at room temperature, while the conductivity in the SrRuO₃ layer is dominant at lower temperature. Supplementary Fig. 3i shows temperature dependence of magnetization in the same set of the samples. The magnetization at 2 K in SRO5/SIO5/Sub has almost the same value with those of the other samples, suggesting nearly the same thickness of SrRuO₃. Supplementary Figure 3j shows anomalous Hall conductivity (σ_{AHE}) as a function of magnetization (M). The sign of σ_{AHE} in SRO5/SIO5/Sub is reversed roughly at the value of $0.6 \mu_B/\text{Ru}$, which is close to those in the other samples as indicated by arrows in the inset of Supplementary Fig. 3j.

The influence of the thicker SrIrO₃ is also observed in V_G dependence of ρ_{xx} as well as the suppressed electrical control of AHE mentioned in the main text. Supplementary Figures 4a-d show longitudinal resistivity variations $[\rho_{xx} - \rho_{xx}(0 \text{ V})]/\rho_{xx}(0 \text{ V})$ as a function of V_G at low temperatures. In the thinner SrIrO₃ sample (SRO5/SIO2/Sub, Supplementary Fig. 4b), the V_G dependence shows the behaviour of electron systems, which is the same with that in SRO5/Sub

(Supplementary Fig. 4a). By contrast, in the thicker SrIrO₃ sample (SRO5/SIO5/Sub, Supplementary Fig. 4c), the V_G dependence shows the behaviour of hole systems, which is the same with that in SIO5/Sub (Supplementary Fig. 4d). The change in modulated carrier type indicates that most of the electric field is screened within the SrIrO₃ layer to give the hole-type carrier density modulation, resulting in the smaller electric-field effect to the SRO/SIO interface in SRO5/SIO5/Sub. This is consistent with the smaller variation in anomalous Hall resistivity as shown in Fig. 2f in the main text.

Supplementary Note 4 | Magneto-transport properties in all heterostructures

In Supplementary Figs. 5a-g, we show the sum of anomalous and topological Hall resistivities ($\rho_{\text{AHE}} + \rho_{\text{THE}}$) at various temperatures as a function of magnetic field in all examined heterostructures, where the thickness of SrRuO₃ ranges from 3 to 5 unit cells. In this thickness region of SrRuO₃, the heterostructures exhibit topological Hall effect (THE) as an additional term deviated from the conventional ferromagnetic hysteresis curve of AHE regardless of the stacking sequence of SrRuO₃ and SrIrO₃. Both the sign of AHE and the appearance of THE in SIO2/SRO_m/Sub are qualitatively the same with the previous report⁴ dealing with the same stacking order. We note that AHE with inverted structures (SRO_m/SIO2/Sub) indicates a slight difference from the reported structures (SIO2/SRO_m/Sub). When we look at 4-unit-cell SrRuO₃ samples (SRO4/SIO2/Sub and SIO2/SRO4/Sub), for example, the sign inversion of AHE occurs in SRO4/SIO2/Sub while it does not in SIO2/SRO4/Sub, exhibiting positive sign of the AHE in all measured temperatures. This difference is probably ascribed to the slight discordance of chemical potential or electron localization, which may come from some distortions in crystals depending on the stacking sequence.

Supplementary Note 5 | Negligible T_C variation by applied electric field

In Supplementary Figs. 6a and 6b, we show temperature dependences of ρ_{xx} and $d\rho_{xx}/dT$ under different electric fields in SRO5/SIO2/Sub. Since the ferromagnetic transition in SrRuO₃ is accompanied by a kink structure of ρ_{xx} , we can estimate the electric-field-induced T_C variation from the anomaly in ρ_{xx} , which is observed as the increase in $d\rho_{xx}/dT$. However, the observed $d\rho_{xx}/dT$ is almost unchanged by electric field, suggesting the negligible shift of T_C . This V_G -independent behaviour of T_C implies that the ferromagnetic interaction J is almost unvaried by the applied electric field because the change of J should contribute to the shift of T_C .

Supplementary Note 6 | Basic transport properties in SrRuO₃/SrIrO₃/SrTiO₃

In Supplementary Figs. 7a-c, we show basic transport properties in SRO m /SIO2/Sub ($m = 3, 4$, and 5 unit cells). It is known that high-quality SrRuO₃ films on atomically flat SrTiO₃(001) substrates exhibit metallic conduction and ferromagnetism until the thickness is reduced to approximately 3 unit cells^{5,6}. However, it is not straightforward to grow such a thin itinerant ferromagnetic SrRuO₃ layer on a SrIrO₃ layer, even though the 2-unit-cell SrIrO₃ seems atomically flat according to its surface morphology as shown in Supplementary Fig. 3b. By carefully optimizing growth conditions, our heterostructures composed of SrRuO₃ and SrIrO₃ maintain their itinerant ferromagnetic temperature dependences (Supplementary Fig. 7a) until the thickness of SrRuO₃ reaches the thin limit of 3 unit cells.

When SrRuO₃ layer thickness is reduced, the longitudinal resistivity is enhanced (Supplementary Fig. 7a). At the same time, ferromagnetic Curie temperature and magnetization normal to the film plane are suppressed compared with those of bulk⁷

($T_C \sim 160$ K, $M \sim 1.6 \mu_B/\text{Ru}$) as shown in Supplementary Fig. 7b. These systematic suppressions of the itinerant ferromagnetic natures are attributed to electron localization in SrRuO_3 and are also observed in other ultrathin SrRuO_3 films^{5,6}. As a result, anomalous Hall conductivity (σ_{AHE}) shows no sign inversion in $\text{SRO}_3/\text{SIO}_2/\text{Sub}$ (Supplementary Fig. 7c), which is consistent with the scaling character of σ_{AHE} as a function of M . The thin SrIrO_3 layer likely contributes to electron conduction, while the 2-unit-cell SrIrO_3 alone appears to be insulating with a resistivity over $10 \Omega \text{ cm}$ at room temperature.

Supplementary Note 7 | Topological Hall effect in $\text{SrRuO}_3/\text{SrIrO}_3/\text{SrTiO}_3$

Finite THE is extracted from magneto-transport ($\rho_{\text{AHE}} + \rho_{\text{THE}}$) and MOKE measurements in $\text{SRO}_m/\text{SIO}_2/\text{Sub}$ ($m = 3$ and 4) (Supplementary Figs. 8a and 8b) as well as that in $m = 5$ (Supplementary Fig. 8c) as already shown in Fig. 2b in the main text. In all measurements, the magnetic fields at which hysteresis curves are closed in MOKE measurements approximately coincide with those in magneto-transport measurements. This indicates that the both measurements were properly performed at almost the same temperature. Kerr rotations normalized by $\rho_{\text{AHE}} + \rho_{\text{THE}}$ at those magnetic fields represent the anomalous Hall component because both measurements represent only ferromagnetic magnetization. At higher temperatures in $\text{SRO}_3/\text{SIO}_2/\text{Sub}$, however, the magneto-transport properties cease to represent pure AHE and THE because the nature of multicarrier in SrRuO_3 appears in Hall effect near T_C . As a result, we were not able to deduce the reliable value of THE in this sample at 50 K and more.

Supplementary Note 8 | Temperature dependence of electrical modulations in SrRuO₃/SrIrO₃/SrTiO₃

In Supplementary Figs. 9a-e, we show the series of magneto-transport properties and their electric-field modulations from which we deduce Fig. 4 in the main text. Since ρ_{xx} is inversely proportional to carrier density (n), the observed $\Delta\rho_{xx}/\rho_{xx}$ (0 V) is consistent with the expected electron accumulation by a SrTiO₃ back-gate transistor. At 10 K, for example, the measured electron density was $2 \times 10^{15} \text{ cm}^{-2}$ ($1 \times 10^{22} \text{ cm}^{-3}$ in 5 unit cells of SrRuO₃) under $V_G = 0 \text{ V}$. Meanwhile, the accumulated sheet carrier density is expected to be about $1 \times 10^{13} \text{ cm}^{-2}$ under $V_G = 200 \text{ V}$, which corresponds to 0.5% decrease in ρ_{xx} , that is, approximately the half value of the observed difference between plus (200 V) and minus (−180 V) gate bias. Therefore, we were roughly able to regard $\Delta\rho_{xx}/\rho_{xx}$ (0 V) as the modulation of carrier density. In all measurements of the electric-field effect, all of the transport properties (ρ_{xx} , ρ_{AHE} , and ρ_{THE}) under $V_G = 0 \text{ V}$ are located between those under $V_G = 200 \text{ V}$ and $V_G = -180 \text{ V}$. The ρ_{THE} gets larger as the thickness of SrRuO₃ gets thinner as shown in Supplementary Fig. 9d. This thickness dependence of ρ_{THE} is consistent with the previous report in the structure of SIO₂/SRO_m/Sub⁴.

Supplementary References

1. Gao, R. *et al.* Interfacial Octahedral Rotation Mismatch Control of the Symmetry and Properties of SrRuO₃. *ACS Appl. Mater. Interfaces* **8**, 14871–14878 (2016).
2. Wu, F.-X. *et al.* Metal-insulator transition in SrIrO₃ with strong spin-orbit interaction. *J. Phys. Condens. Matter* **25**, 125604 (2013).
3. Matsuno, J. *et al.* Engineering a Spin-Orbital Magnetic Insulator by Tailoring Superlattices. *Phys. Rev. Lett.* **114**, 247209 (2015).
4. Matsuno, J. *et al.* Interface-driven topological Hall effect in SrRuO₃-SrIrO₃ bilayer. *Sci. Adv.* **2**, e1600304 (2016).
5. Xia, J., Siemons, W., Koster, G., Beasley, M. R. & Kapitulnik, A. Critical thickness for itinerant ferromagnetism in ultrathin films of SrRuO₃. *Phys. Rev. B* **79**, 140407(R) (2009).
6. Chang, Y. J. *et al.* Fundamental thickness limit of itinerant ferromagnetic SrRuO₃ thin films. *Phys. Rev. Lett.* **103**, 057201 (2009).
7. Cao, G., McCall, S., Shepard, M., Crow, J. E. & Guertin, R. P. Thermal, magnetic, and transport properties of single-crystal Sr_{1-x}Ca_xRuO₃ (0<x<1.0). *Phys. Rev. B* **56**, 321–329 (1997).