

[Supplementary Information]

Tunable magnons of an antiferromagnetic Mott insulator via interfacial metal-insulator transitions

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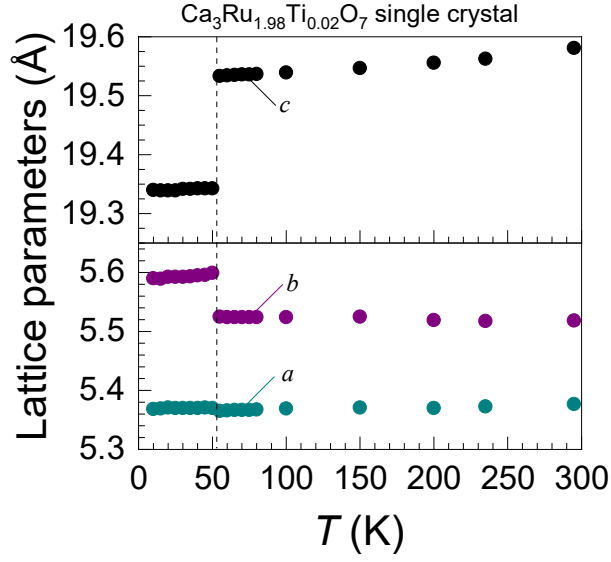
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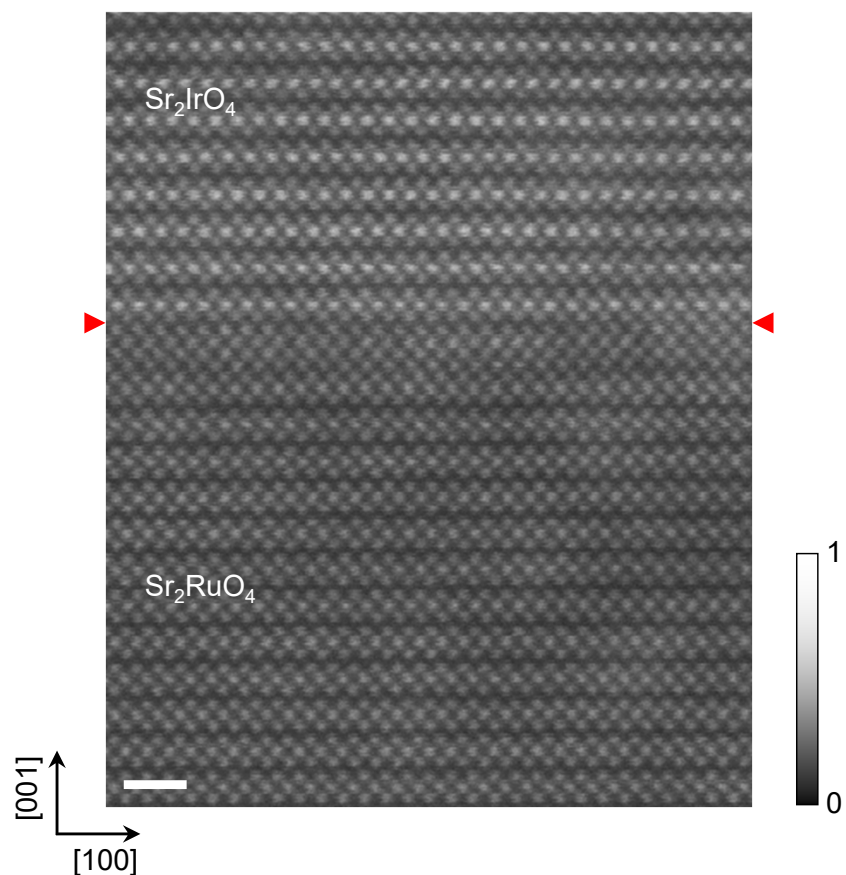
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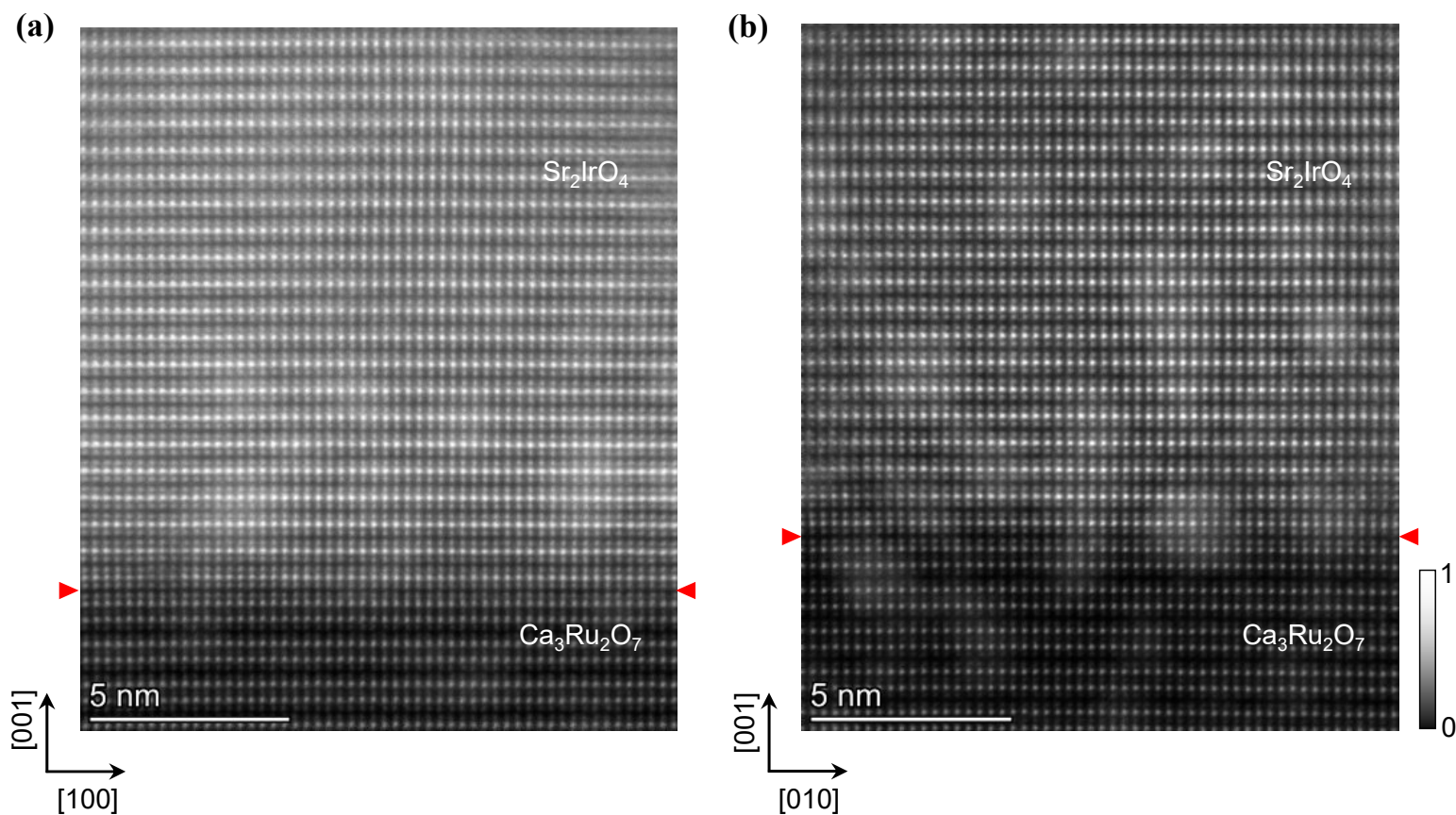
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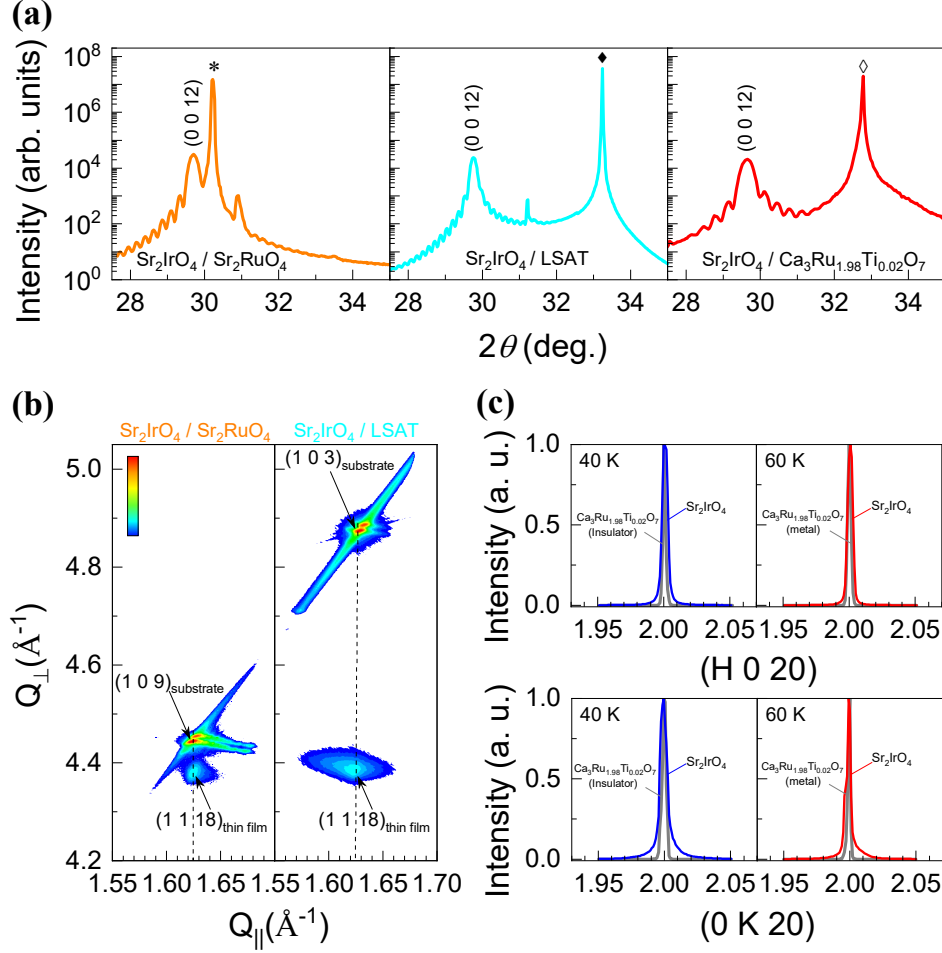
Supplementary Figure 1. The temperature-dependent lattice constants of our Ca₃Ru_{1.98}Ti_{0.02}O₇ single crystal reveal distinct changes in the b and c axes at 55 K. This temperature corresponds to a metal-insulator transition: below 55 K, the sample exhibits insulating behavior, whereas it remains metallic above this temperature.



Supplementary Figure 2. A cross-sectional Z-contrast scanning transmission electron microscopy (STEM) image of a $\text{Sr}_2\text{IrO}_4/\text{Sr}_2\text{RuO}_4$ heterostructure showing a single-phase Sr_2IrO_4 thin film epitaxially grown on a Sr_2RuO_4 single crystal. Red triangles mark the atomically sharp interface between Sr_2IrO_4 and Sr_2RuO_4 . Scale bar: 1 nm.



Supplementary Figure 3. Cross-sectional Z-contrast STEM images of a $\text{Sr}_2\text{IrO}_4/\text{Ca}_3\text{Ru}_2\text{O}_7$ heterostructure taken along (a) the $[100]$ and (b) the $[010]$ crystallographic directions. The images reveal a single-phase Sr_2IrO_4 thin film with an atomically sharp interface to the $\text{Ca}_3\text{Ru}_2\text{O}_7$ substrate. Red triangles indicate the position of the interface.



Supplementary Figure 4. (a) X-ray diffraction patterns of $\text{Sr}_2\text{IrO}_4/\text{Sr}_2\text{RuO}_4$, $\text{Sr}_2\text{IrO}_4/\text{LSAT}$, and $\text{Sr}_2\text{IrO}_4/\text{Ca}_3\text{Ru}_{1.98}\text{Ti}_{0.02}\text{O}_7$ heterostructures, showing $(0\ 0\ l)$ diffraction peaks of the Sr_2IrO_4 thin films with thickness fringes. The estimated film thicknesses are ~ 30 nm for $\text{Sr}_2\text{IrO}_4/\text{Sr}_2\text{RuO}_4$, ~ 50 nm for $\text{Sr}_2\text{IrO}_4/\text{LSAT}$, and ~ 20 nm for $\text{Sr}_2\text{IrO}_4/\text{Ca}_3\text{Ru}_{1.98}\text{Ti}_{0.02}\text{O}_7$. The small peak observed in both scans arises from interference effects and is not related to the Sr_2IrO_4 crystal structure. (b) Reciprocal space maps around the $(1\ 0\ 9)$ reflection of the Sr_2RuO_4 substrate and the $(1\ 0\ 3)$ reflection of the LSAT substrate, clearly show $(1\ 1\ 18)$ -reflection of the Sr_2IrO_4 thin films. These maps confirm that the Sr_2IrO_4 films are fully strained to both substrates, exhibiting a compressive strain of approximately 0.51%. The intensity is displayed on a logarithmic scale. (c) H- and K-scans of the $\text{Sr}_2\text{IrO}_4/\text{Ca}_3\text{Ru}_{1.98}\text{Ti}_{0.02}\text{O}_7$ heterostructure acquired at temperatures above and below 55 K—the temperature at which the substrate undergoes a metal-insulator transition. In both temperature regimes, the Sr_2IrO_4 thin film remains fully strained to the substrate. The H- and K-scans were performed near the $(2\ 0\ 16)$ and $(0\ 2\ 16)$ reflections of the $\text{Ca}_3\text{Ru}_{1.98}\text{Ti}_{0.02}\text{O}_7$ substrate and the corresponding $(2\ 0\ 20)$ and $(0\ 2\ 20)$ reflections of the Sr_2IrO_4 thin films.

	a (Å)	c (Å)	Lattice mismatch (%)	ϵ_{xx} (%)
Sr ₂ IrO ₄ single crystal	3.89 (Orthorhombic: 5.50)	25.78 ($c/2 = 12.89$)		
Sr ₂ RuO ₄ single crystal	3.87	12.73	-0.51	
LSAT single crystal	3.87	3.87	-0.51	
Sr ₂ IrO ₄ / Sr ₂ RuO ₄	3.87 ± 0.01 (Orthorhombic: 5.47)	25.914 ± 0.002 ($c/2 = 12.96$)		-0.51 ± 0.26
Sr ₂ IrO ₄ / LSAT	3.87 ± 0.02 (Orthorhombic: 5.49)	25.89 ± 0.02 ($c/2 = 12.95$)		-0.51 ± 0.41

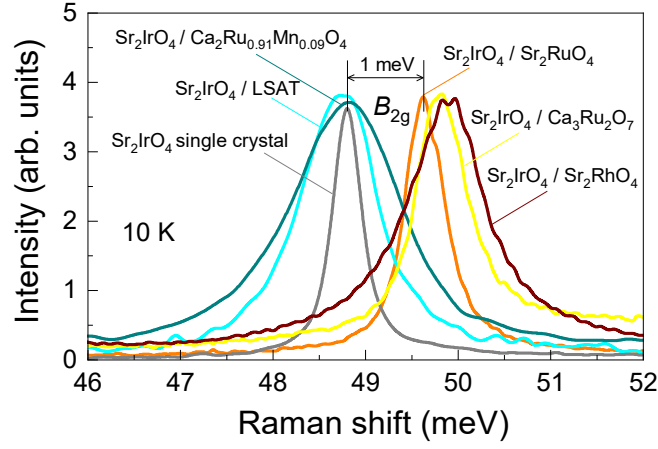
Supplementary Table 1. In-plane and out-of-plane lattice parameters of Sr₂IrO₄ thin films, Sr₂IrO₄ single crystal, Sr₂RuO₄, and LSAT single crystals. Lattice mismatch values between Sr₂IrO₄ and the substrates are calculated using the formula: $((a_{\text{sub}} - a_{\text{bulk}})/a_{\text{sub}}) \times 100$ (%). In-plane misfit strain values of the Sr₂IrO₄ thin films are calculated as: $((a_{\text{film}} - a_{\text{bulk}})/a_{\text{bulk}}) \times 100$ (%). Error bars represent uncertainties estimated from the instrumental accuracy.

	a (Å)	b (Å)	c (Å)	ε_{xx} (%)	ε_{yy} (%)	ε_{zz} (%)
Sr ₂ IrO ₄ crystal (295 K)	5.499	5.499	25.784	-	-	-
Sr ₂ IrO ₄ crystal (13 K)	5.485	5.485	25.798	-	-	-
Sr ₂ IrO ₄ / Ca ₃ Ru _{1.98} Ti _{0.02} O ₇ (295 K)	5.377(1)	5.517(1)	25.96(2)	-2.22(2)	+0.33(2)	+0.70(5)
Sr ₂ IrO ₄ / Ca ₃ Ru _{1.98} Ti _{0.02} O ₇ (6 K)	5.365(1)	5.594(1)	25.81(2)	-2.19(2)	+1.99(2)	+0.06(5)

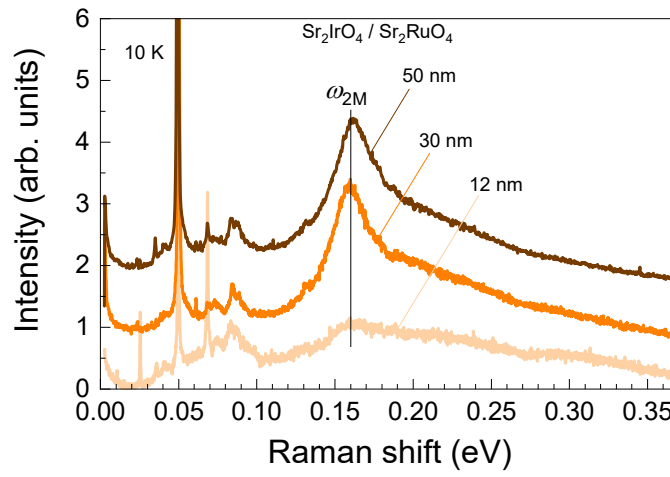
Supplementary Table 2. In-plane and out-of-plane lattice parameters of the Sr₂IrO₄/Ca₃Ru_{1.98}Ti_{0.02}O₇ heterostructure measured at room temperature and at 6 K. Upon cooling to 6 K, the tensile strain along the b -axis increases, indicating enhanced lattice distortion.

	J_1 (meV)	J_2 (meV)	J_3 (meV)	J_4 (meV)
Sr ₂ IrO ₄ single crystal	54	-17	16	7.7
Sr ₂ IrO ₄ /LSAT	55	-17	16	7.3
Sr ₂ IrO ₄ /Sr ₂ RuO ₄	55	-21	20	4.6

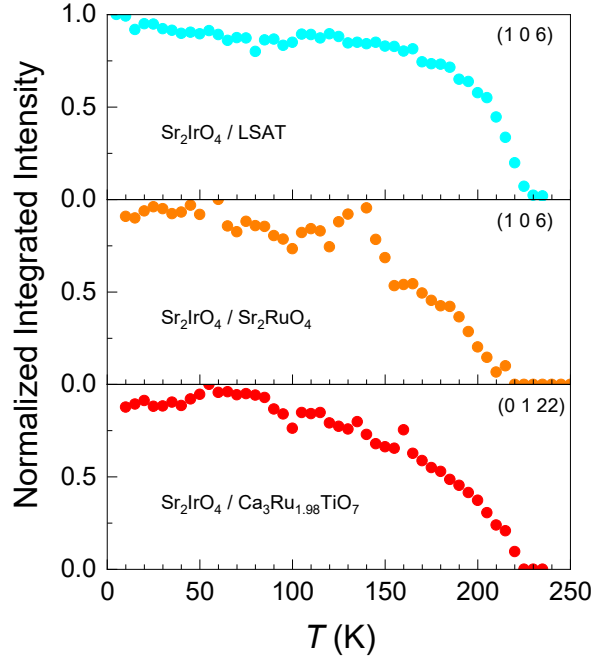
Supplementary Table 3. Comparison of exchange interaction parameters for Sr₂IrO₄/Sr₂RuO₄, Sr₂IrO₄/LSAT, and Sr₂IrO₄ single crystal, obtained by fitting the magnon dispersion data using a model Hamiltonian that includes first-, second-, third-, fourth-nearest neighbor exchange interactions (denoted as J_1 , J_2 , J_3 , and J_4 , respectively).



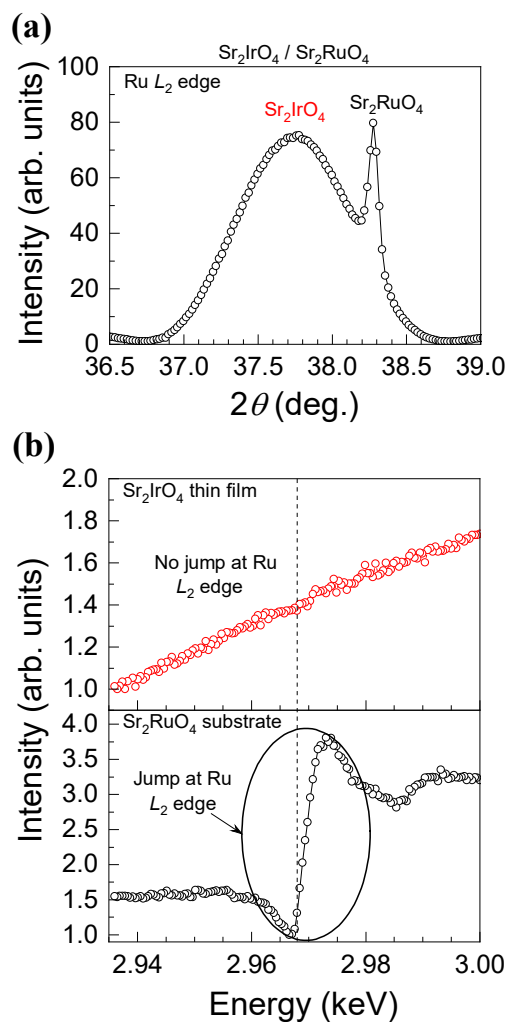
Supplementary Figure 5. B_{2g} phonon mode of Sr_2IrO_4 thin films deposited on various substrates. The data clearly show that Sr_2IrO_4 thin films grown on metallic substrates exhibit a hardened B_{2g} phonon mode, whereas films deposited on insulating substrates display phonon energies similar to that of the Sr_2IrO_4 single crystal.



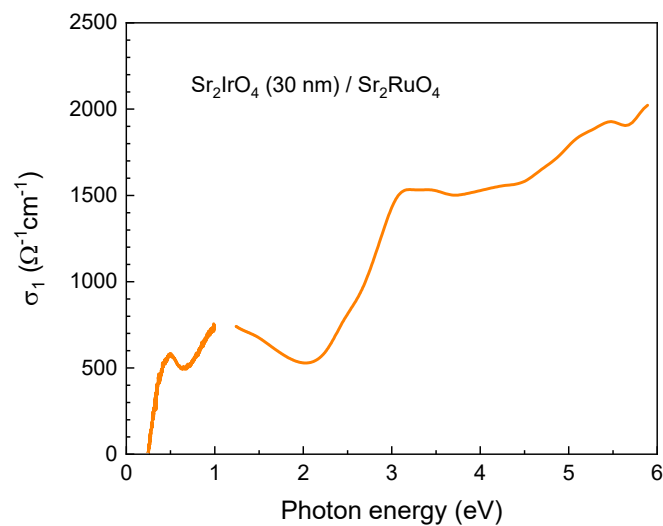
Supplementary Figure 6. Thickness-dependent Raman spectra of the $\text{Sr}_2\text{IrO}_4/\text{Sr}_2\text{RuO}_4$ heterostructure measured at 10 K, showing no significant variation in the two-magnon peak energy with film thickness.



Supplementary Figure 7. Temperature dependence of the integrated intensity of the Ir L_3 -edge magnetic Bragg peaks: the $(1\ 0\ 6)$ peak for Sr_2IrO_4 thin films deposited on Sr_2RuO_4 and LSAT substrates, and the $(0\ 1\ 22)$ peak for the $\text{Sr}_2\text{IrO}_4/\text{Ca}_3\text{Ru}_{1.98}\text{Ti}_{0.02}\text{O}_7$ heterostructure. No significant shift in the Néel temperature is observed for Sr_2IrO_4 thin films across these different substrate conditions.



Supplementary Figure 8. (a) 2θ scan of the $\text{Sr}_2\text{IrO}_4/\text{Sr}_2\text{RuO}_4$ heterostructure performed at the Ru L_2 absorption edge. (b) Energy scan across the Ru L_2 edge measured from both the Sr_2IrO_4 thin film side and the Sr_2RuO_4 substrate near the interface. The absence of an absorption jump on the Sr_2IrO_4 side confirms that no detectable Ru impurities are present in the Sr_2IrO_4 thin film.



Supplementary Figure 9. Optical conductivity spectra of a 30-nm-thick Sr₂IrO₄ thin film grown on a Sr₂RuO₄ substrate, measured at room temperature.