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SUPPLEMENTARY INFORMATION
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Ultrafast energy- and momentum-resolved dynamics of magnetic correlations in the photo-doped Mott insulator Sr_2IrO_4

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Here we present details of the sample synthesis and characterization and further information about the optical reflectivity measurements.

SAMPLE SYNTHESIS AND CHARACTERIZATION

The Sr_2IrO_4 thin film sample was produced using pulsed laser deposition [1]. A KrF excimer laser was used to ablate a stoichiometric Sr_2IrO_4 target 5.5 cm away from the sample with 1.1 J/cm^2 fluence pulses at a 1 Hz repetition frequency. For the growth, the $SrTiO_3$ substrate was held at 850°C in 1 mTorr of background oxygen pressure. After growth the films were cooled down in 1 atmosphere of oxygen pressure. Figure S1 plots an $(0, 0, L)$ X-ray diffraction measurement of the film, taken with a laboratory Cu $K\alpha$ x-ray source. Strong Bragg peaks are visible from the film and the substrate. Within the precision of the measurement, no impurity phases were detected. Figure S2 presents X-ray characterization of the film taken at SACLA. Good crystallinity is shown in the $(0, 0, 28)$ and $(-2, -2, 24)$ structural Bragg peak rocking curves (Fig. S2a,b), with full-width at half-maximum mosaics of 0.10° and 0.18° respectively. The energy scan about the Ir L_3-edge, shown in panel (c), shows a strong white line resonance, as seen in previous work [2].

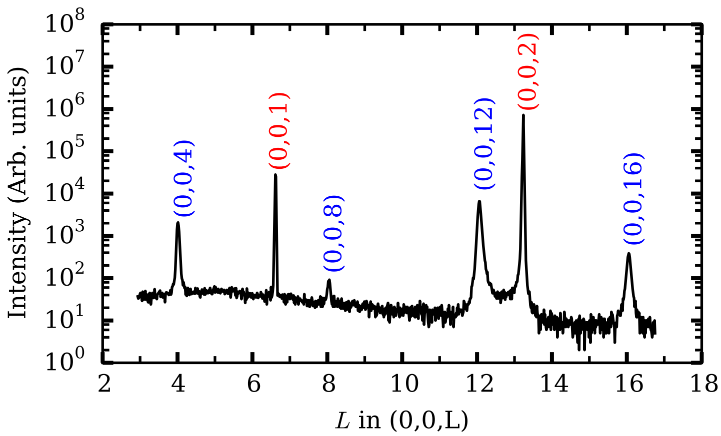

FIG. S1. $(0, 0, L)$ diffraction measurement on the Sr_2IrO_4 thin film. All peaks can be accounted for without any impurity phases. Bragg peaks from Sr_2IrO_4 and $SrTiO_3$ are indexed in blue and red respectively. L in the x-axis is defined in terms of the Sr_2IrO_4 lattice with $c = 25.83$ Å.

The single crystal Sr_2IrO_4 sample was prepared from $SrCO_3$, IrO_2, and $SrCl_2$ starting materials with a molar ratio of 1.8:1.0:15. $SrCl_2$ acted as a flux. The mixture was melted at 1300°C and subsequently cooled down to 900°C at a rate of 8°C per hour before being furnace-cooled to room temperature. The Néel temperature was determined from bulk magnetization in a magnetic field

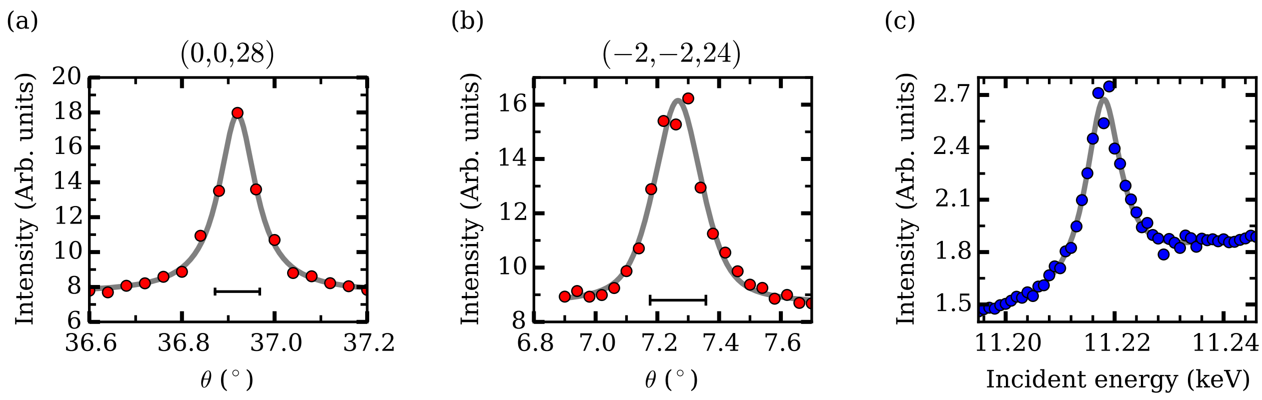

FIG. S2. Diffraction measurements of the Sr_2IrO_4 thin film taken at SACLA showing good quality crystallinity. (a) and (b) plot the $(0, 0, 28)$ and $(-2, -2, 24)$ structural peaks respectively. Horizontal bars show the full-width at half-maximum of the peaks. (c) X-ray fluorescence measurement as a function of incident energy through the Ir L_3-edge resonance.

of 0.5 T. Further details of the sample characterization are described in Ref. [3].

OPTICAL REFLECTIVITY MEASUREMENTS

The transient changes in the Sr_2IrO_4 optical reflectivity were measured in a standard optical pump-probe setup, on the same single crystal that was used in the RIXS measurements. The 2 μm, 100-fs excitation pulses were derived from the idler beam of a single-stage optical parametric amplifier, pumped by a 1 kHz Ti:sapphire amplifier system that also provides the 800-nm probe pulses. Excitation fluences in the mJ/cm^2 range were achieved by focusing the pump beam to a spot size of about 300 μm. The sample was held at 110 K base temperature using a continuous flow cryostat.

Figure 2d in the main text plots the normalized change in optical reflectivity as a function of pump fluence. The recovery of the normalized change in reflectivity, $\Delta R(t)/R$, can be fit with two exponential terms as

$$\frac{\Delta R(t)}{R} = -A_{\text{fast}} \exp(-t/T_{\text{fast}}) - A_{\text{slow}} \exp(-t/T_{\text{slow}}) \tag{1}$$

where T_{fast} and T_{slow} are the fast and slow recovery timescales and A_{fast} and A_{slow} are the respective amplitudes of these processes. This formula was fit to the recovery data starting 300 fs after the pump.

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