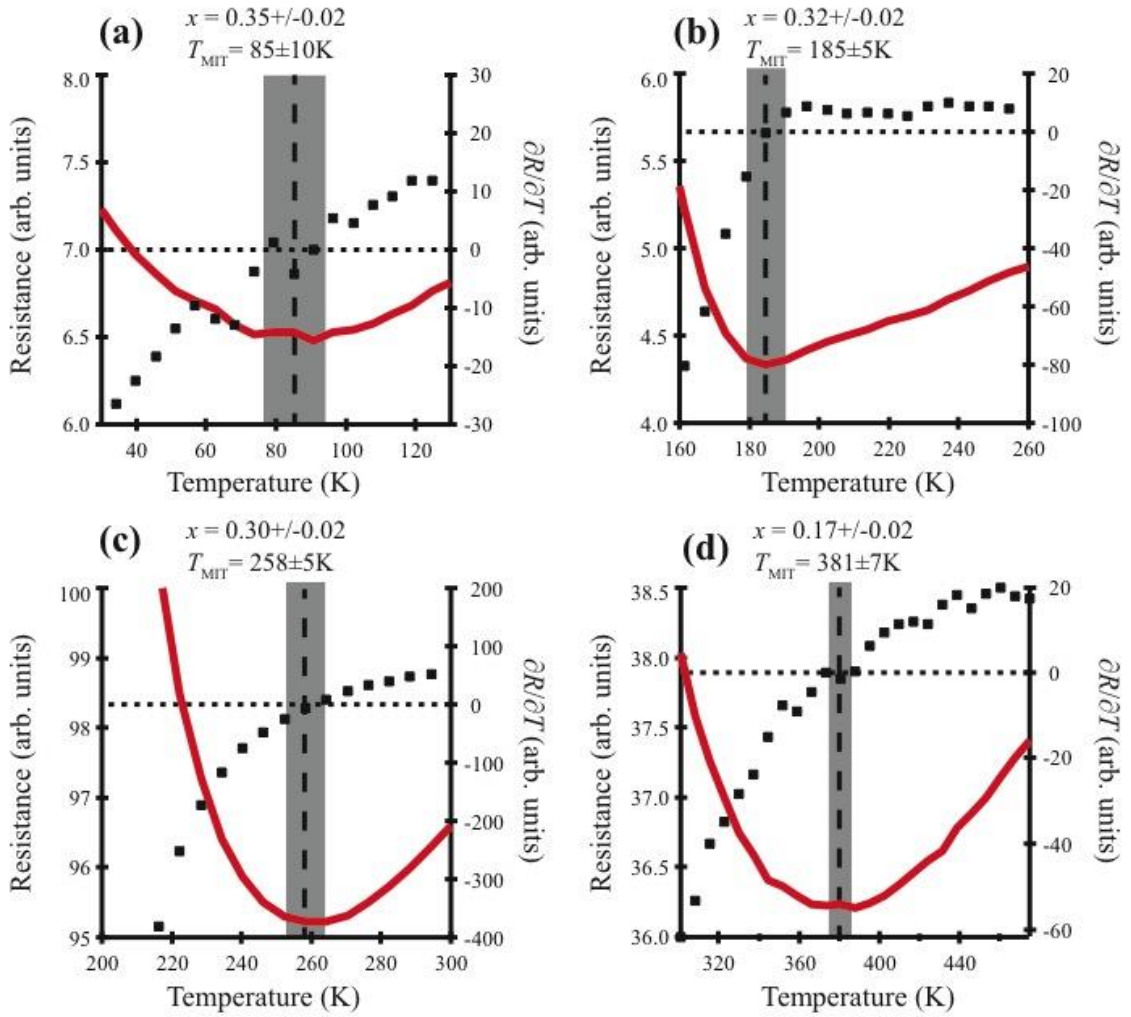
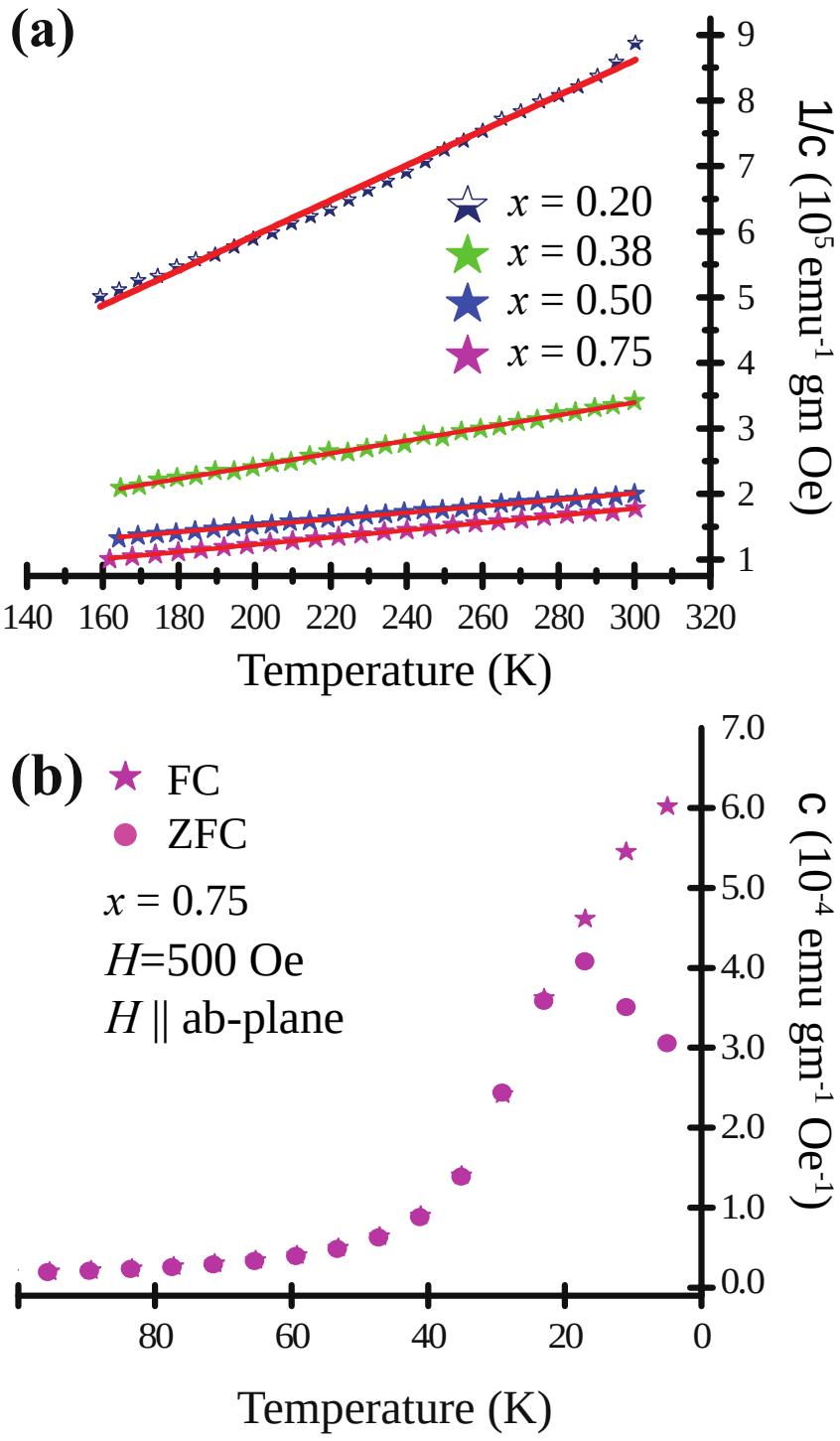
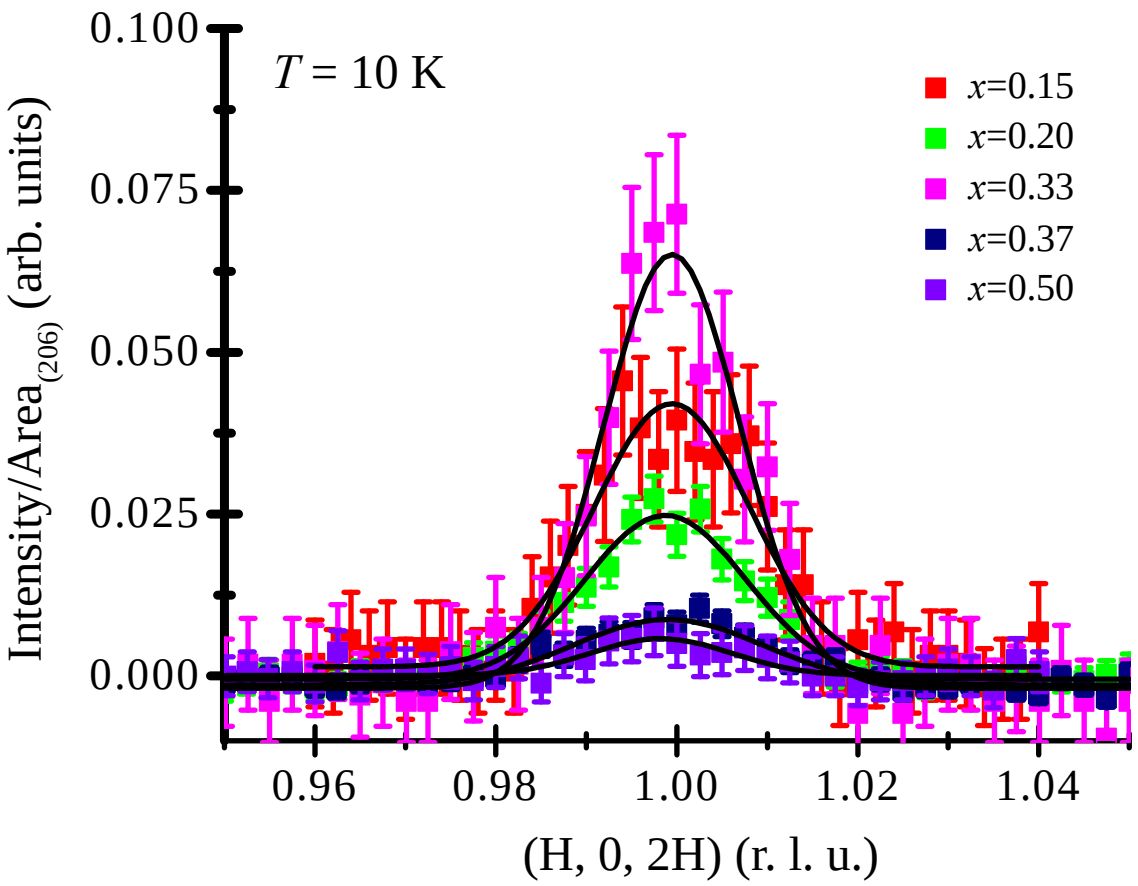
Supplementary Figures:



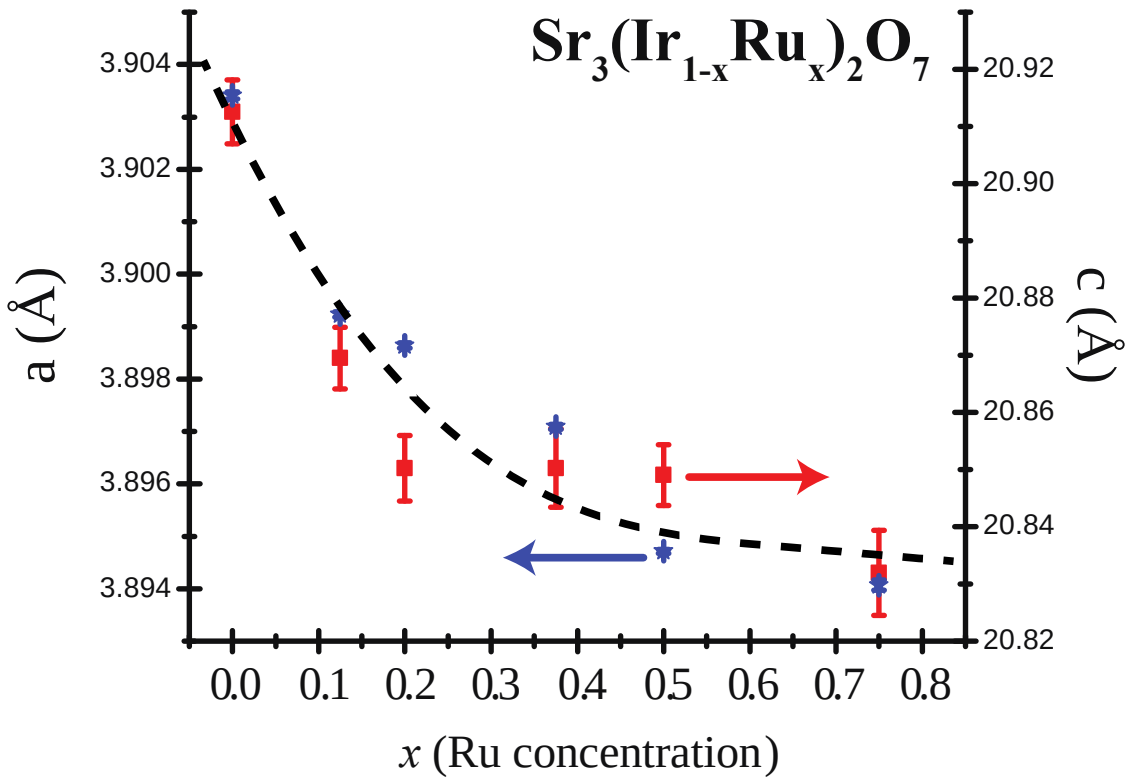
Supplementary Figure 1: Resistance (red lines) and its first derivative $\delta R/\delta T$ plotted (black squares) plotted as a function of temperature for $Sr_3(Ir_{1-x}Ru_x)_2O_7$ samples with (a) x=0.35, (b) x=0.32, (c) x=0.3, and (d) x=0.17 respectively. Vertical dashed lines show the temperature T_{MIT} plotted in the electronic phase diagram of Fig. 1 (a) in the paper's main text. Shaded grey region shows the uncertainty in determining T_{MIT}. Ru x-concentrations were measured for each sample with the corresponding uncertainty displayed in each panel. Resistance and $\delta R/\delta T$ are plotted as raw data in arbitrary units without geometric conversion to resistivity.



Supplementary Figure 2: (a) χ^{-1} as a function plotted as a function of temperature. A 500 Oe field was applied parallel to the ab-plane for all concentrations with the exception of x=0.2 where a 1 T field was applied. Solid lines are fits to Curie-Weiss behavior. (b) Zero-field cooled (ZFC) and field cooled (FC) magnetization data collected for the x=0.75 concentration. 1 emu g^{-1} Oe^{-1} = 4π x 10^{-3} m^3/kg.



Supplementary Figure 3: Radial scans through the antiferromagnetic Bragg reflection **Q**=(1,0,2) for select concentrations of $Sr_3(Ir_{1-x}Ru_x)_2O_7$. Intensity for each sample has been divided by the integrated area of the sample's corresponding **Q**=(2,0,6) nuclear Bragg reflection. Before correcting for minor changes in absorption and extinction between Ru-concentrations this plot provides a rough illustration of the moment evolution as a function of *x*. Solid lines are Gaussian fits to the data. Error bars are one standard deviation. An additional sample, not plotted here, with nominal *x*=0.32 was measured with a normalized scattering intensity that saturates at 0.3 (off the scale) and the corresponding AF moment plotted in Fig. 3 (c) of the main text.



Supplementary Figure 4: Lattice parameters determined via powder x-ray diffraction on crushed crystals plotted as a function of Ru-doping for $Sr_3(Ir_{1-x}Ru_x)_2O_7$ at 300K. Data was refine using the FullProf Rietveld refinement program[1]. Dashed line is a guide to the eye. While the reduction in lattice constants is monotonic, it is not linear and seemingly maps the nonlinear contraction previously observed in $Sr_2(Ir_{1-x}Ru_x)O_4$.[2] Error bars are one standard deviation.

Supplementary References:

[1] J. Rodriguez-Carvajal, Recent Advances in Magnetic Structure Determination by Neutron Powder Diffraction, Physica B192, 55 (1993).

[2] R. J. Cava, B. Batlogg, K. Kiyono, H. Takagi, J. J. Krajewski, W. F. Peck, Jr., L. W. Rupp, Jr., and C. H. Chen, Localized-to-itinerant electron transition in $Sr_2Ir_{1-x}Ru_xO_4$**,** Phys. Rev. B 49, 11890 (1994).