

Supplemental Material

Evolution of Magnetism in Single-Crystal $\text{Ca}_2\text{Ru}_{1-x}\text{Ir}_x\text{O}_4$ ($0 \leq x \leq 0.65$)

S. J. Yuan^{1*}, J. Terzic¹, J. C. Wang^{1,2,3}, L. Li¹, S. Aswartham¹, W. H. Song^{1,4}, F. Ye², and G. Cao^{1*}

¹*Center for Advanced Materials, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506, USA*

²*Quantum Condensed Matter Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

³*Department of Physics, Renmin University of China, Beijing 100872, China*

⁴*Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, China*

Crystal Structure Refinement

More than 3000 reflections (500 + unique reflections) were collected for each single crystal during single-crystal XRD experiments.

The high quality of our refinement is evidenced in the following parameters:

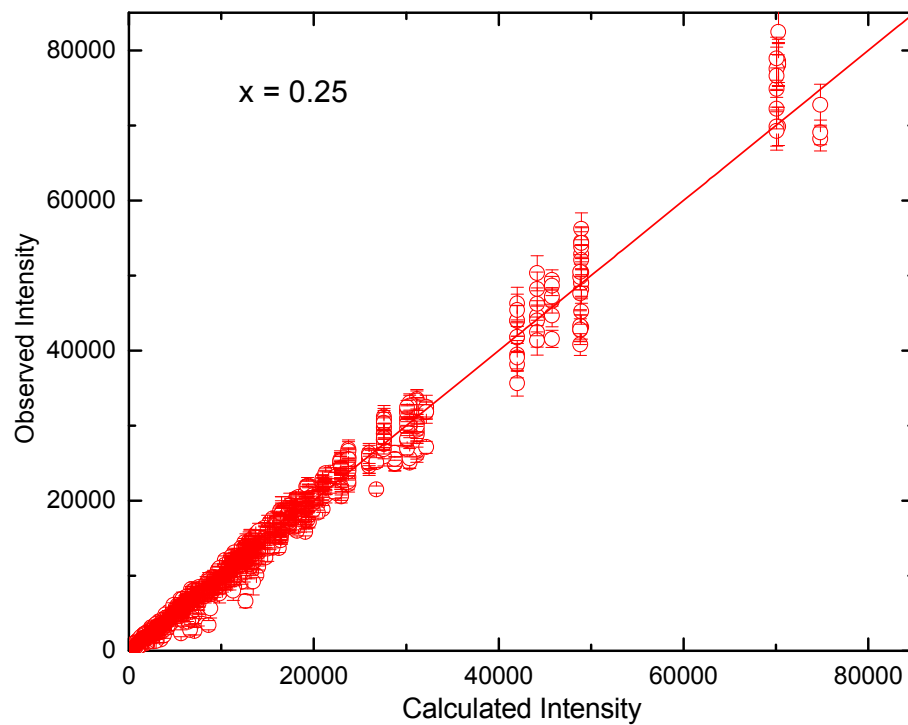
Goodness of fit = 1.113

R1 = 0.0160

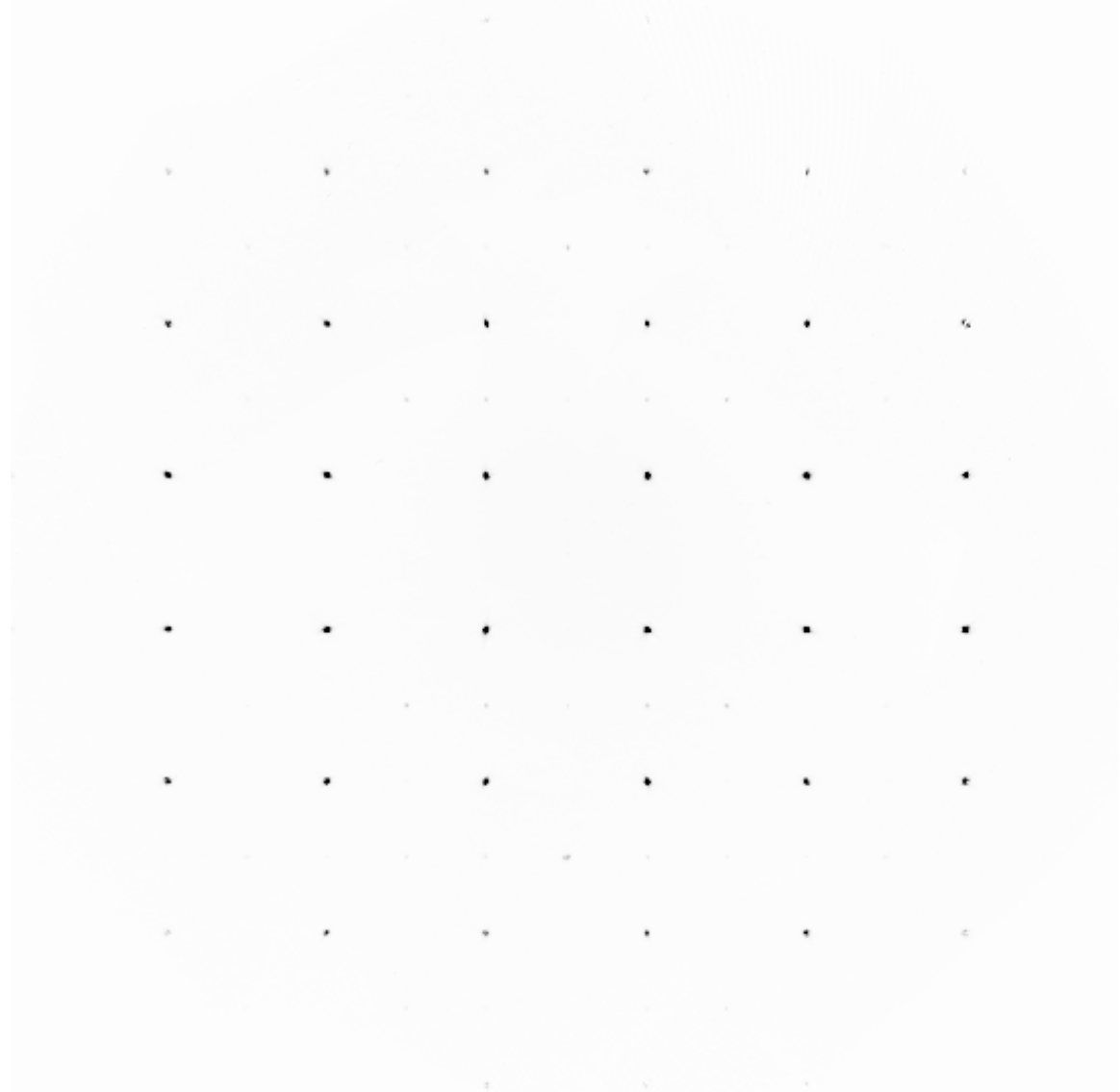
wR2 = 0.0443

The high-quality refinement is also corroborated in **SM Fig. 1**, in which Observed Intensity as a function of Calculated Intensity is presented. Such a plot is often used as a measure of the quality of refinement. In addition, the high quality of the single crystals is also clearly illustrated in the x-ray diffraction pattern in **MS Fig. 2**. The sharp dots indicate a highly ordered crystals structure.

All results indicate that the refined parameters presented in the manuscript (atomic positions, occupations, thermal displacements, etc.) are unambiguously reliable and that the single crystals studied are of high quality.



SM Fig.1. A representative plot of Observed Intensity as a function of Calculated Intensity for $x=0.25$



SM Fig. 2. A representative diffraction pattern $[hk3]$ for single crystal $\text{CaRu}_{1-x}\text{Ir}_x\text{O}_4$.