Mapping the fluctuating $\text{Ir}^{4+}$ dimers across the phase diagram of $\text{Cu(Ir}_{1-x}\text{Cr}_x\text{)}_2\text{S}_4$ ($0 \leq x \leq 0.6$)\textsuperscript{1} E.S. BOZIN, Brookhaven National Laboratory, Y.S. HOR, J.F. MITCHELL, Argonne National Laboratory, S.J.L. BILLINGE, P. JUHAS, Brookhaven National Laboratory, Columbia University — Elucidating the role of fluctuations in systems with competing interactions, such as $\text{Cu(Ir}_{1-x}\text{Cr}_x\text{)}_2\text{S}_4$, enables comprehensive understanding of their physical properties. While $\text{CuIr}_2\text{S}_4$ exhibits complex behavior with metal-insulator transition accompanied by charge ordering and $\text{Ir}^{4+}$-spin-dimerization at 226 K, $\text{CuCr}_2\text{S}_4$ displays ferromagnetic metallic behavior ($T_C \sim 377$ K) understood within the double exchange model. Intermediate composition range sees suppression of end-member properties, with broad features observed in susceptibility around 180 K attributed to $\text{Cr}^{3+}$ low spin to high-spin crossover [1]. Robust fluctuating $\text{Ir}^{4+}$ dimers are detected and their evolution examined across the phase diagram by the X-ray atomic pair distribution function method. Although their long range order is destroyed already by $x \approx 0.05$, $\text{Ir}^{4+}$ dimers exist locally at low temperature at all compositions studied. We provide detailed account of the Cr-doping and temperature dependence of the local dimers, and estimate characteristic length-scale on which they are observable. Fluctuating dimers disappear on heating, for intermediate compositions at temperatures above 180 K.

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