\( J_{\text{eff}} = 1/2 \) Mott spin-orbit insulating state close to the cubic limit in \( \text{Ca}_4\text{IrO}_6 \)


The \( J_{\text{eff}} = 1/2 \) Mott spin-orbit insulating state is manifested in systems with large cubic crystal field splitting and spin-orbit coupling that are comparable to the on-site Coulomb interaction, \( U \). 5d transition metal oxides host parameters in this regime and recently strong evidence for this state in \( \text{Sr}_2\text{IrO}_4 \), and additional iridates, has been presented. All the candidates, however, have distorted octahedra, such as the elongation along the c-axis in \( \text{Sr}_2\text{IrO}_4 \), and consequently a non-cubic local crystal field environment. Consequently the materials form a mixed \( J_{\text{eff}} = 1/2, 3/2 \) ground state. The lack of a material with an unmixed \( J_{\text{eff}} = 1/2 \) has impacted the development and testing of robust models of this novel insulating and magnetic state. We present neutron diffraction, resonant x-ray scattering and DFT calculations that not only reveal \( \text{Ca}_4\text{IrO}_6 \) is a new candidate \( J_{\text{eff}} = 1/2 \) material with long-range magnetic order, but furthermore resides close to the required cubic limit. Both our experimental and theoretical investigation indicate \( \text{Ca}_4\text{IrO}_6 \) is uniquely positioned to act as a canonical system to investigate of the \( J_{\text{eff}} = 1/2 \) state.

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