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Title: Pressure-driven collapse of the relativistic electronic ground state in a honeycomb iridate

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Abstract:

Honeycomb-lattice quantum magnets with strong spin-orbit coupling are promising candidates for realizing a Kitaev quantum spin liquid. Although iridate materials such as Li2IrO3 and Na2IrO3 have been extensively investigated in this context, there is still considerable debate as to whether a localized relativistic wavefunction (Jeff = 1/2) provides a suitable description for the electronic ground state of these materials. To address this question, we have studied the evolution of the structural and electronic properties of α -Li2IrO3 as a function of applied hydrostatic pressure using a combination of x-ray diffraction and x-ray spectroscopy techniques. We observe striking changes even under the application of only small hydrostatic pressure ($P \le 0.1$ GPa): a distortion of the Ir honeycomb lattice (via X-ray diffraction), a dramatic decrease in the strength of spin-orbit coupling effects (via X-ray absorption spectroscopy), and a significant increase in non-cubic crystal electric field splitting (via resonant inelastic X-ray scattering). Our data indicate that α -Li2IrO3 is best described by a Jeff = 1/2 state at ambient pressure, but demonstrate that this state is extremely fragile and collapses under the influence of applied pressure.

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