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Pressure-induced collapse of the $J_{\text{eff}} = 1/2$ ground state in Li_2IrO_3

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The honeycomb lattice iridate Li_2IrO_3 displays a novel $J_{\text{eff}} = 1/2$ Mott insulating ground state driven by strong 5d spin-orbit coupling effects. Due to a combination of $J_{\text{eff}} = 1/2$ magnetic moments, 90 degree Ir-O-Ir bond geometry, and honeycomb lattice crystal structure, Li_2IrO_3 represents one of the most promising candidates for the experimental realization of the Kitaev model. This exactly solvable quantum spin model features highly anisotropic, bond-dependent magnetic interactions, and supports an exotic spin liquid ground state. Although the observation of long-range magnetic order ($T_n \sim 15$ K) excludes a “pure” Kitaev model description of Li_2IrO_3 , there are many “extended” Kitaev models (including contributions such as isotropic Heisenberg exchange, further-neighbor interactions, symmetric off-diagonal exchange, and structural distortions) that may be relevant to this material. As such, there is considerable interest in potential strategies for “tuning” Li_2IrO_3 through the use of external perturbations.

We have employed a combination of x-ray powder diffraction (XPD), resonant inelastic x-ray scattering (RIXS), and x-ray absorption spectroscopy (XAS) techniques to investigate how the structural, electronic, and magnetic properties of Li_2IrO_3 evolve as a function of applied pressure. We find evidence of a pressure-induced structural phase transition at $P \sim 3$ GPa, which is accompanied by a dramatic increase in the non-cubic crystal electric field splitting. Furthermore, we observe a rapid drop in the XAS branching ratio, indicating that applied pressure drives Li_2IrO_3 out of the strong spin-orbit regime and leads to a collapse of the $J_{\text{eff}} = 1/2$ ground state.

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