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Pressure-induced collapse of the J_eff = 1/2 ground state in Li2IrO3

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The honeycomb lattice iridate Li2IrO3 displays a novel J_eff = 1/2 Mott insulating ground state driven by strong 5d spin-orbit coupling effects. Due to a combination of J_eff = 1/2 magnetic moments, 90 degree Ir-O-Ir bond geometry, and honeycomb lattice crystal structure, Li2IrO3 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 (Tn ~ 15 K) excludes a "pure"Kitaev model description of Li2IrO3, 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" Li2IrO3 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 Li2IrO3 evolve as a function of applied pressure. We find evidence of a pressure-induced structural phase transition at P ˜ 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 Li2IrO3 out of the strong spin-orbit regime and leads to a collapse of the J_eff = 1/2 ground state.

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