Evolution of Novel Magnetism in Single-Crystal Honeycomb Iridates: An Approach towards Quantum Spin Liquids

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Iridium-based oxides have become a fertile ground for novel physics driven by strong spin-orbit coupling. Among them, honeycomb lattices Na_2IrO_3 and Li_2IrO_3 have drawn a great deal of attention because a unique combination of the crystal structure and strong spin-orbit coupling these materials adopt provides a new paradigm for studies of exotic states such as the long sought-after quantum spin liquid and topological quantum phase transition. Procedural methods included a self flux-method for single-crystal synthesis, structural characterizations through X-Ray diffraction methods and energy dispersion X-Ray analyses, and physical property measurements (electrical resistivity, magnetization, and specific heat). This work reports the successful synthesis of single-crystals of the layered iridate, $(Na_{1-x}Li_x)_2IrO_3$, and a thorough study of its structural, magnetic, thermal and transport properties. The new compound allows a controlled interpolation between Na_2IrO_3 and Li_2IrO_3, while maintaining the novel quantum magnetism of the honeycomb Ir^{4+} planes. The measured phase diagram demonstrates a suppression of the Néel temperature, T_N, at an intermediate x indicating that the magnetic order in Na_2IrO_3 and Li_2IrO_3 are distinct. At x = 0.70, T_N is most suppressed and the honeycomb structure is least distorted; this suggests that this compound is closest to the spin liquid that is elusive in Na_2IrO_3 and Li_2IrO_3. Ultimately, this approach towards a new state of matter – the quantum spin liquid – could lead to exponential improvements in data storage, long-range communication, high temperature superconductivity, and quantum computation.

Awards Won:

Third Award of $1,000