

Abstract

Quantum spin liquids (QSLs) are exotic states of matter, in which magnetic frustration and strong quantum fluctuations destroy long-range magnetic order. Highly frustrated lattices with antiferromagnetic exchange interactions and low-spin value open up possibilities in the exploration and designing of new QSL candidates. QSLs in three-dimensions are very rare, and only a couple of candidate materials exist.

In this thesis, we investigate bond-disordered quantum spin-liquid state in three dimensional magnetic insulators $\text{Na}_4\text{Ir}_3\text{O}_8$ and $\text{Ca}_{10}\text{Cr}_7\text{O}_{28}$. The central theme of thesis is to explore how these QSL's respond to external perturbations like pressure, magnetic field, and chemical substitutions. In this thesis, I have synthesized two QSL candidates the hyperkagome $\text{Na}_4\text{Ir}_3\text{O}_8$ and kagome bilayer $\text{Ca}_{10}\text{Cr}_7\text{O}_{28}$ and studied their electrical transport, magnetic, and thermal properties. Our work provides several new results: (i) the strongly frustrated Mott insulating state in $\text{Na}_4\text{Ir}_3\text{O}_8$ is quite robust against large removal of Na from the lattice, (ii) evidence through magnetic (Ru) and nonmagnetic (Ti) impurity doping in a magnetic sublattice, of fragile magnetic order and importance of nearestneighbour interactions and spin-orbit coupling in deciding the magnetic ground state in $\text{Na}_4\text{Ir}_3\text{O}_8$, (iii) high-pressure magnetic susceptibility measurements reveal the QSL at ambient pressure is quite robust and may not depend on a delicate balance between any specific values of competing exchange interactions in $\text{Ca}_{10}\text{Cr}_7\text{O}_{28}$, and (iv) the first experimental realization of a perfect nonmagnetic analog $\text{Ca}_{10}\text{V}_7\text{O}_{27.5}$ of $\text{Ca}_{10}\text{Cr}_7\text{O}_{28}$, which enables an accurate exclusion of the lattice heat capacities.