

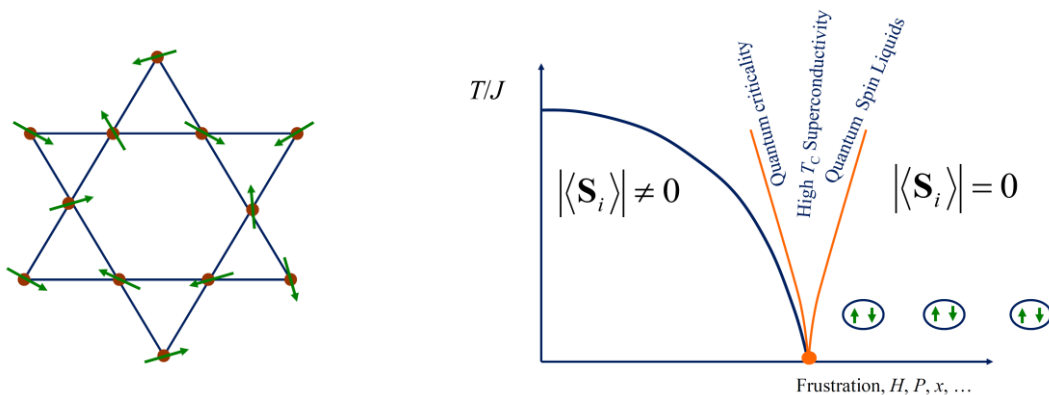
Competition-induced Novel Quantum States

Correlations, competing interactions, and quantum character are major ingredients which are at the basis of most of the ground breaking discoveries in hard condensed matter that brought new scientific concepts and challenges as well as promising applications (high temperature superconductivity in cuprates, high thermoelectric power in cobaltates, multiferroics...).

The focus of this proposal was to explore even more exotic states of matter- so-called emergent phases- with not yet identified properties and ground states which result from **competition**. Competition can be taken in a broad sense, competition between close in energy ground states in the vicinity of a quantum critical point, competition at the microscopic level such as generated by frustration. Frustration is at the heart of much science, from protein folding to emergent electromagnetism, but reaches its purest manifestation in magnetic materials where it refers to the presence of competing interactions that can't be simultaneously satisfied at variance with more common ferro- or antiferro-magnets where organized states are stabilized. Married with quantum fluctuations, frustration can indeed give birth to exotic states such as a quantum "spin liquid" (QSL) a well identified case in one dimension but which has been a Grail quest in two and three dimensions since the 70s –the RVB conjecture for triangular-based antiferromagnetic lattices [1].

In recent years some novel frustration-induced spin liquids have been synthesized enabling a one to one long-sought confrontation between theoretical models and experiments. Three inspiring tracks for this field of research have been the seed of our experimental work on QSL (i) revealing ultimate properties of state of the art materials, this is the case of the emblematic kagome compound $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ known under its mineral name, herbertsmithite; (ii) deepen our understanding on more recently discovered QSL; (iii) explore new tracks issued from novel materials synthesis marrying frustration and a complex interplay between spin, orbital, and charge degrees of freedom [2].

From a pure experimental point of view, the current research proposal focused on the investigation of the microscopic static magnetism and low energy excitations of these collective quantum states by nuclear magnetic resonance (NMR) and muon spin relaxation (μSR) accompanied by advanced sub-Kelvin thermodynamic and magnetization studies on frustrated magnets. We provide a brief summary on each of the materials studied during the course of the project which belong to three different classes Herbertsmithite kagome, hyperkagome and spin-orbit driven triangular lattice antiferromagnets



Left: Geometrical frustration on a corner sharing triangular lattice named "kagome" In reference to a popular weaving of baskets in Japan. Right: Quantum Phase transition and emergent phenomena in condensed matter.

Kagome Herbertsmithite: $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

Herbersmithite on a kagome lattice is one of the model quantum spin liquid candidates, wherein strong frustration induced quantum fluctuations melts any kind of ordering down to mK. It was a daunting task to pin-point the exact shift intrinsic to the kagome network and the role of Cu/Zn defects given the broad NMR spectra in polycrystalline materials. Taking advantage of the strong coupling of O to the Cu kagome network, we have carried out comprehensive ^{17}O NMR on high quality single crystals of Herbersmithite and tracked the static and dynamic magnetic susceptibilities intrinsic to the kagome lattice down to 1.4 K along two orientations. While one cannot get rid completely of the Cu/Zn defects signature in NMR spectra, the different spectral signatures from Cu spins on the kagome lattice and those on Zn site offer the opportunity for the discussion of the nature of spin excitation spectrum of the ground-state. The defects contribution at low- T is clearly singled out and has been studied in great detail. The temperature dependence of static and dynamic susceptibilities point towards a gapless quantum spin liquid state in the kagome Herbersmithite [3]. Torque and ESR measurements reveal a breaking of symmetry at low temperatures [4].

2. Spin Liquid state in the 3D frustrated Hyperkagome : $\text{PbCuTe}_2\text{O}_6$

The novel quantum magnet $\text{PbCuTe}_2\text{O}_6$ wherein frustration arises due to specific so-called “hyperkagome” magnetic lattice, made of corner-sharing triangles of Cu^{2+} ions constituting a 3D structure. The magnetization and specific heat down to 300 mK don’t show any signature of long range magnetic ordering except a small anomaly at 0.86 K attributed to the presence of a tiny fraction of defects in the polycrystalline sample. The local spin susceptibility tracked by the NMR shift hardly deviates from the macroscopic one down to 1 K pointing to a homogeneous magnetic system with a low defect concentration. The saturation of the NMR shift and the sub-linear power law T -evolution of the $1/T_1$ - NMR relaxation rate at low T demonstrate a non-singlet ground state favoring a gapless fermionic description of the low energy magnetic excitations. Below 1 K a pronounced slowing down of the spin dynamics is witnessed both by the μSR relaxation and the wipe out of the NMR intensity, and may correspond to an instability of the spinon Fermi surface. $\text{PbCuTe}_2\text{O}_6$ represents the rare example of a 3D spin liquid candidate with $S = \frac{1}{2}$ [5].

3. Disordered ground state in spin-orbit driven Ir based triangular lattice $\text{Ba}_3\text{InIr}_2\text{O}_9$

Our preliminary T -dependence of magnetization data on Ir-based magnet on a frustrated triangular compound, $\text{Ba}_3\text{InIr}_2\text{O}_9$, a candidate for the Heisenberg-Kitaev model shows a bifurcation of ZFC-FC at 1 K in small magnetic fields. This suggests the presence of a tiny fraction of defect spins/grain boundaries in the polycrystalline sample. The specific heat data do not show any signature of magnetic ordering down to 300 mK suggesting a disordered ground state. NMR measurements are underway to get insights into the intrinsic magnetic susceptibilities and spin dynamics inherent to the magnetic lattice. μSR will shed light on the microscopic coexistence or separation of magnetic phases or evidencing minute impurity phases with distinct magnetic signatures [6].

References

[1] P. W. Anderson Science **235**, 1196 (1987). [2] L. Balents, Nature **464**, 199 (2010) [3] **P. Khuntia**, P. Mendels, F. Bert *et al.*, (in preparation). [4] A. Zorko, M. Herak, M. Gomilsek, J. van Tol, M. Velazquez, **P. Khuntia**, F. Bert, and P. Mendels. [5] **P. Khuntia**, F. Bert, P. Mendels, B. Koteswararao, A. V. Mahajan, M. Baenitz, F. C. Chou, C. Baines, A. Amato, and Y. Furukawa, Phys. Rev. Lett. **116**, 107203 (2016). [6] **P. Khuntia et al.**, (unpublished).