Experimental report

14/02/2018

Proposal: **4-05-662** **Council:** 10/2016

Title: Magnetic excitation spectrum of a potential spin liquid PbCuTe2O6

Research area: Physics

This proposal is a new proposal

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Samples: PbCuTe2O6

Instrument	Requested days	Allocated days	From	To
THALES	6	6	24/01/2017	30/01/2017

Abstract:

PbCuTe2O6 consists of a 3-dimensional arrangement of spin-1/2 Cu ions. There are 3 potentially strong interactions, the first neighbor interaction couples the magnetic ions into isolated triangles, the second couples them into the highly frustrated hyperkagome lattice consisting of corner sharing triangles and the third neighbor interaction couples them into antiferromagnetic chains. In PbCuTe2O6 the hyperkagome interaction dominates suppressing long-range magnetic order down to 20 mK making it a promising candidate to look for spin liquid ground state. We wish to explore the magnetic excitations of this quantum magnet using inelastic neutron scattering on the THALES spectrometer.

Fractional excitations in a potential quantum spin liquid

Unlike conventional magnets where the magnetic moments are partially or completely static in the ground state, in a spin liquid they remain in collective motion down to the lowest temperatures. The importance of this state is that it is coherent and highly entangled without breaking local symmetries. Such phenomena is usually sought in lattices where anisotropies, favouring specific spin alignments, are incompatible with the lattice geometries and interactions between spins as is the case for the spin ice compounds. In Heisenberg spin systems where anisotropies are absent, spin liquid behaviour can arise when the moments form a network of elementary motifs such as triangles or tetrahedra, and interact with each other via antiferromagnetic interactions. In some cases it is impossible to simultaneously satisfy all the exchange interactions and the competition between them can leads to exotic ground states where long-range magnetic order is suppressed to low temperatures or completely eliminated. In three dimensions there are many well-known experimental examples of networks of corner-shared tetrahedra, particularly in the pyrochlore and spinel structures. On the other hand, *three-dimensional networks of corner-sharing triangles* are mostly unexplored, despite the expectation that they would also display novel ground states. In this experiment we have investigated the magnetic excitations of a potential spin liquid compound $PbCuTe_2O_6$ which consists of a *frustrated three dimensional network of corner-sharing triangles* made up of spin-1/2 magnetic ions.

$PbCuTe_2O_6$ crystallizes in cubic structure with lattice constant a=12.4454 Å [1]. The positions of the magnetic Cu^{2+} ions in $PbCuTe_2O_6$ are shown in Figure 1a where the green and red bonds represent the 1st and 2nd nearest neighbour interactions J1 and J2, respectively. On its own J1 couples the Cu^{2+} moments into isolated triangles, while J2 forms a three-dimensional network of corner-sharing triangles known as the hyperkagome lattice. Further neighbour interactions J3, J4 (shown in figure 1b) form isolated chains running parallel to the crystalline a, b, c axes and parallel to the body diagonals respectively. It is known from DC susceptibility and MuSR results that the system does not order down to 0.02K [2,3]. Curie-Weiss temperature (= -22K) from DC susceptibility and density functional theory calculations reveal that all four interactions of $PbCuTe_2O_6$ are antiferromagnetic in nature and their ratio is found to be J1:J2:J3:J4= 1.06:1:0.55:0.11, indicating that the system is strongly frustrated making it a potential quantum spin liquid candidate [4].

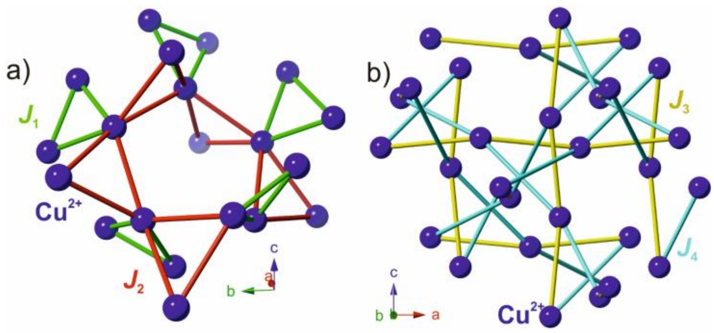

Figure 1: a) The three dimensional triangular network of Cu^{2+} ions formed by the 1st and 2nd nearest neighbour interactions in PbCuTe2O6 and b) shows the 3rd neighbours forming 1D chains running along the crystalline a,b,,c axes and 4th neighbors form chains parallel to the body diagonals.

The fundamental excitations of a spin liquid are spin-1/2 excitations known as spinons. Experimentally, these are observed in inelastic neutron scattering as broad and diffuse features rather than the sharp modes typical to magnons or spin-waves. The spinon excitations of the kagome (corner-sharing triangles in 2-dimensions) spin liquid Herbertsmithite formed diffuse ring-like features which have been successfully measured and characterized. Similar observation has been made in other spin liquid candidates such as $CaCr_{10}O_8$ and alpha-RuCl3. Our preliminary measurements on the powder sample of $PbCuTe_2O_6$ reveal the presence of such diffuse magnetic excitations originating at $|Q|$~0.8$Å^{-1}$ [4]. The excitation band is dispersionless and extends up to 3meV. In the current experiment we have mapped out momentum-resolved distribution of the magnetic excitation spectrum in a single crystal of $PbCuTe_2O_6$ using inelastic neutron scattering at ThALES.

The crystal was aligned in <100>/<010> scattering plane and the excitation map was measured at temperatures 2K and below 0.1K for energy transfers between 0.25 meV to 2meV. Figure 2 shows the <hk0> maps at the energy transfer of 0.5 meV at the two temperatures. At both temperatures, the excitations form a diffuse ring at $|Q| \approx 0.8$ Å^{-1}, while additional weaker branches of scattering extend outwards to higher wavevectors. For energies smaller than 1meV at T=40mK, the diffuse ring has double maxima at wavevectors (1.69,~±0.3,0) and (~±0.3,1.69,0), etc (see figure 2a) while at higher energies it broadens and becomes weaker. At T=2K, in figure 2b, the diffuse feature of the excitations is retained however, the doublet feature is not distinct anymore. Furthermore, we observe that the overall intensity is higher than at low temperatures. However, the origin of this difference is not clearly understood yet. Further neutron scattering experiments are required to understand this intensity dependence on temperature and the nature of QSL state itself in $PbCuTe_2O_6$.

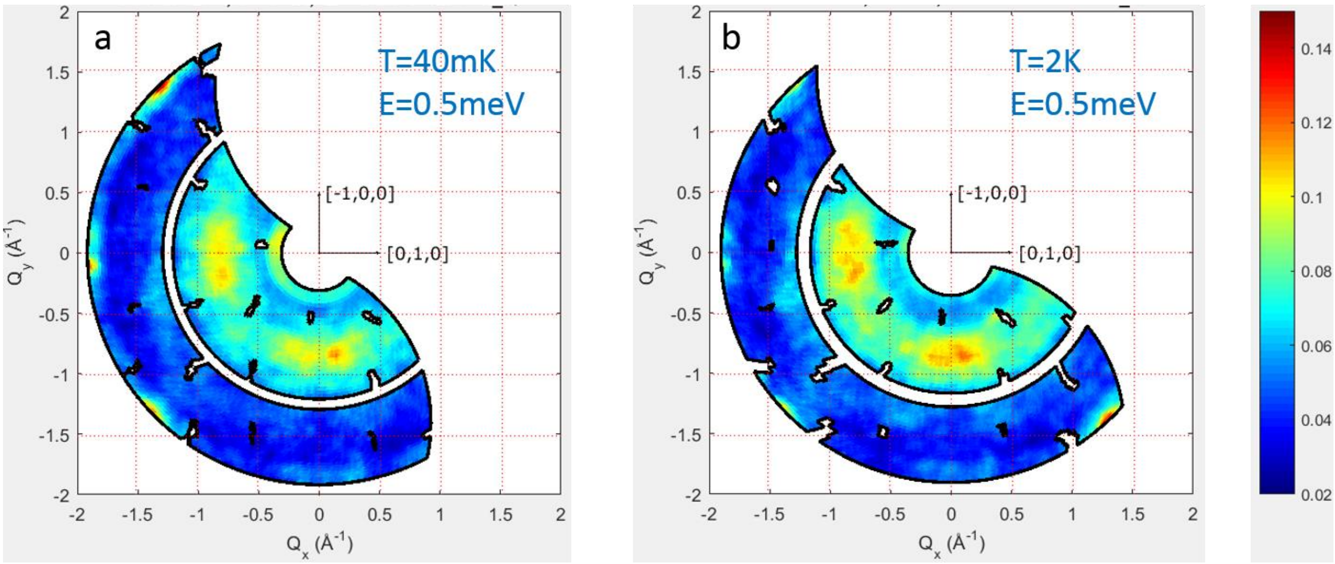

Figure 2: a) The inelastic spectrum of PbCuTe2O6 at 0.1K for an energy transfer of 0.5meV measured at ThALES and b) shows the theoretical static spin susceptibility distribution calculated by PFFRG method.

In conclusion, the excitations of $PbCuTe_2O_6$ are clearly very different from the sharp and dispersive spin-wave excitations expected in conventional magnets with long-range magnetically order ground states or from the gapped and dispersive magnon excitations of dimer magnets [7,8]. They are reminiscent of diffuse scattering features observed in several two dimensional quantum spin liquids such as Herbertsmithite and $CaCr_{10}O_8$ [5,6]. Thus, we can confirm from these results that $PbCuTe_2O_6$ is a new spin liquid candidate based on a three dimensional frustrated lattice. These results are now submitted to Nature Communications for publication and the manuscript is currently under review [4].

References:

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